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Measurement of TOF Histogram in High Energy Part of Neutron Spectrum

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An increase of detector's and data acquisition system's performance made it possible for TOF spectrometer to distinguish the initial part of TOF histogram, which corresponds to cascade and fast neutrons.

The work presents experimentally measured initial parts of TOF spectra, obtained using samples-radiators of Au^{197} and Ho^{165} thin patterns as target materials.

The possibility to reconstruct neutron target station's fast neutron spectrum using experimental histogram is discussed.

Measurements were carried out with a channel duration 100 nanoseconds of the data acquisition system, and proton linear accelerator operating with parameters: proton beam duration 250 nanoseconds at half-maximum current amplitude, proton beam energy 267 MeV, pulsed current 0.01 A, flash frequency 50 Hz. TOF spectrometer base 50 meters provided resolution factor: 6 nanoseconds per meter.

1. Practical implementations for initial histogram's part

Already early works [1] were emphasizing, that high energy part of TOF histogram is important not only for total and partial cross sections measurements. Measurement of initial part of spectra allows to study experimentally target station's leakage neutron flux as function of neutron energy. It is own for each subcritical assembly used as proton beam target, which produces neutrons for TOF experiment.

Research program [1] suggested to make the macroscopic neutron TOF experiments for fast breeder reactors and electronuclear breeders, using several changeable proton beam target stations, each with different material and isotope compositions.

Creation of calculation codes, for nuclear energy devices, can rely not only on measurements of microscopic cross sections. Debugging of systems of multi-group constants is possible, basing on making 'macroscopic' experiments with model assemblies of nuclear energy devices. Comparison with calculation of experimental properties of such assemblies (neutron balance, spatial distribution of neutron reactions, averaged by energy spectrum of neutrons – fission, capture, threshold reactions) is base for adjustments and 'customization' of multi group constants. At average heat release in assembly target ~1 kW, detailed measurements of neutron balance are possible using TOF method [1].

2. Example of experimentally measured fast neutron spectrum

Fast neutron spectra, which must be measured, give requirements for performance and resolution factor of the TOF spectrometer. First example is IBR-30 spectrum and flux at 1000 meter TOF base, experimentally measured by Yu.V. Grigoriev [2].

As we can see in Fig.1, IBR-30 spectrum consists of three parts: prompt fission neutrons in groups №1−№6 with low neutron energy border at 0.4 MeV, Fermi spectrum '1/E' in groups №7−№25, and Maxwell spectrum below neutron energy 0.215 eV.

Table 1: IBR-30 Neutron spectrum at 1000 meters TOF base

ABBN-78 Group №	Energy interval	Δ(N)/N	Neutron flux, ($sec^{-1}\cdot cm^{-2}$)
1	10.5 – 6.5 MeV	0.003	5
2	6.5 – 4.0 MeV	0.02	35
3	4.0 – 2.5 MeV	0.04	70
4	2.5 – 1.4 MeV	0.1	173
5	1.4 – 0.8 MeV	0.24	415
6	0.8 – 0.4 MeV	0.06	104
7	0.4 – 0.2 MeV	0.025	43
8	0.2 – 0.1 MeV	0.021	37
9	100 – 46.5 keV	0.023	40
10	46.5 –21.5 keV	0.023	40
11	21.5 – 10 keV	0.023	40
12	10 – 4.65 keV	0.023	40
13	4.65 – 2.15 keV	0.023	40
14	2.15 – 1 keV	0.023	40
15	1 – 0.465 keV	0.023	40
16	465 – 215 eV	0.023	40
17	215 – 100 eV	0.023	40
18	100 – 46.5 eV	0.023	40
19	46.5 – 21.5 eV	0.023	40
20	21.5 – 10 eV	0.023	40
21	10 – 4.65 eV	0.023	40
22	4.65 – 2.15 eV	0.023	40
23	2.15 – 1.0 eV	0.023	40
24	1.0 – 0.465 eV	0.023	40
25	0.465 – 0.215 eV	0.023	40
26	0.215 – 0.001 eV	0.1	174

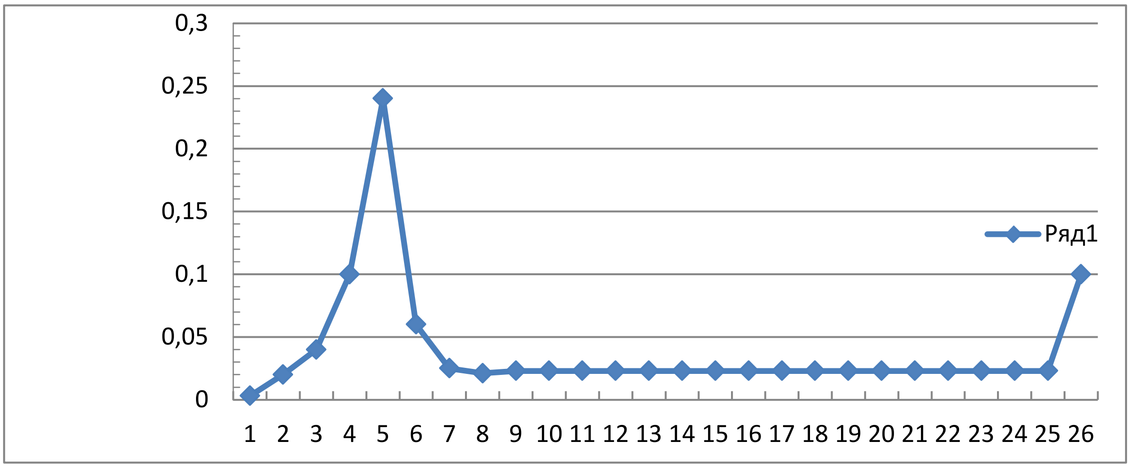

Fig.1. IBR-30 spectrum in energy group intervals of ABBN-78.

3. Numerical modeling results for fast neutron spectra

Example №1 of calculated spectrum: sphere with R=10 cm, made of material: tungsten W(75% vol.) and water H_2O(25% vol.).

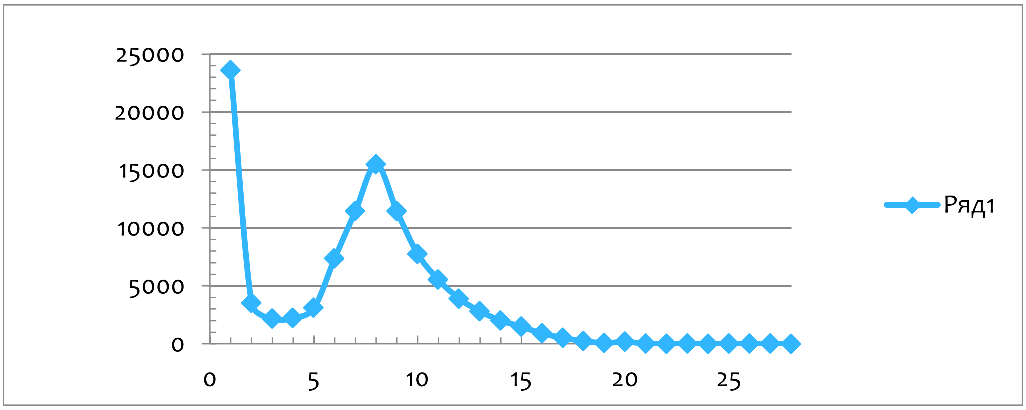

Fig.2. Tungsten-water proton beam target outgoing neutron spectrum.

For calibration of numerical calculation method, neutron spectra of subcritical assemblies were preliminary studied using spherical model with 14 MeV-neutron source in the center.

Using code ‘Shield’ [3,4] for spheres of different substances and different radiuses, were calculated: average leakage neutron spectrum lifetime, spectrum of absorbed neutrons, and spectrum of leakage neutron flux on external surface of the sphere.

For all calculated assemblies, 100,000 neutrons with energy 14 MeV, were placed into the center of the sphere during time t=0. Numbers of neutrons per group are on vertical axis of all figures, and number of ABBN-78 energy group from 1 to 28 on horizontal axis.

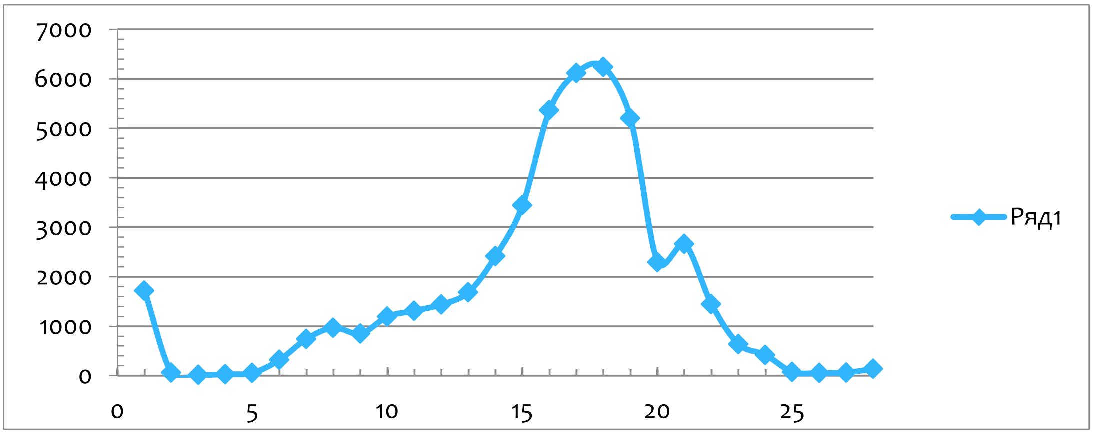

Fig.3. Tungsten-water proton beam target capture neutron spectrum.

As we can see, outgoing spectrum and capture spectrum are two different neutron spectra. They even have maximum number of neutrons in different energy groups.

Example №2 of calculated spectrum: sphere with R=20.1 cm, made of material: U^{235}(20%) and U^{238} (80%).

In this case during t=0 were injected also 100,000 neutrons with energy equal 3 MeV, in ABBN-78 energy group number 3. So as radius was chosen to have small subcriticality of the assembly for chain reaction, resulting number of neutrons is many times bigger than their initial quantity. Majority of neutrons are from fission spectrum. Thus, spectra in this case don’t depend on initial energy of injected neutrons: were found own spectra, typical for assembly’s material composition.

Table 2: Calculation results for W(75% vol) and H_2O(25%) assembly spectra

ABBN-78 Group №	Group № in figures	Energy interval	Outgoing spectrum	Capture spectrum
-1	**1**	14.5 – 14.0 MeV	23586	1721
0	**2**	14.0 – 10.5 MeV	3515	60
1	**3**	10.5 – 6.5 MeV	2159	16
2	**4**	6.5 – 4.0 MeV	2233	29
3	**5**	4.0 – 2.5 MeV	3099	58
4	**6**	2.5 – 1.4 MeV	7362	320
5	**7**	1.4 – 0.8 MeV	11457	745
6	**8**	0.8 – 0.4 MeV	15482	971
7	**9**	0.4 – 0.2 MeV	11442	851
8	**10**	0.2 – 0.1 MeV	7755	1195
9	**11**	100 – 46.5 keV	5526	1313
10	**12**	46.5 – 21.5 keV	3895	1440
11	**13**	21.5 – 10 keV	2804	1689
12	**14**	10 – 4.65 keV	2010	2415
13	**15**	4.65 – 2.15 keV	1477	3446
14	**16**	2.15 – 1 keV	894	5365
15	**17**	1 – 0.465 keV	519	6121
16	**18**	465 – 215 eV	229	6243
17	**19**	215 – 100 eV	79	5208
18	**20**	100 – 46.5 eV	171	2297
19	**21**	46.5 – 21.5 eV	22	2662
20	**22**	21.5 – 10 eV	1	1447
21	**23**	10 – 4.65 eV	17	641
22	**24**	4.65 – 2.15 eV	1	419
23	**25**	2.15 – 1.0 eV	15	76
24	**26**	1.0 – 0.465 eV	14	55
25	**27**	0.465 – 0.215 eV	11	65
26	**28**	0.215 – 0.001 eV	3	137

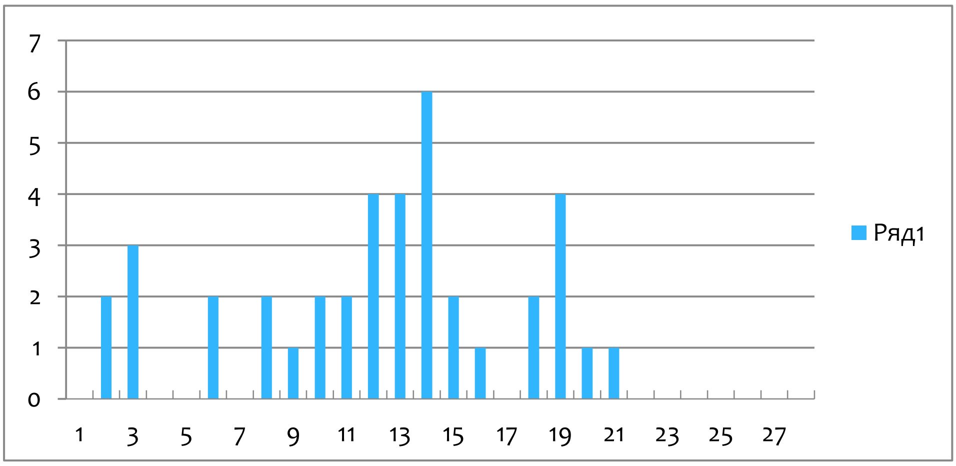

Fig.4. High density LiD sphere R=0.1 cm outgoing spectrum.

Table 3: Calculation results for U^{235}(20%) and U^{238}(20%) assembly spectra

ABBN-78 Group №	Group number in figures	Energy interval	Outgoing spectrum	Capture spectrum
-1	**1**	14.5 – 14.0 MeV	0	0
0	**2**	14.0 – 10.5 MeV	2225	10
1	**3**	10.5 – 6.5 MeV	50112	634
2	**4**	6.5 – 4.0 MeV	247693	6366
3	**5**	4.0 – 2.5 MeV	461785	23193
4	**6**	2.5 – 1.4 MeV	807517	93034
5	**7**	1.4 – 0.8 MeV	1278327	319074
6	**8**	0.8 – 0.4 MeV	2122993	664144
7	**9**	0.4 – 0.2 MeV	1737957	769338
8	**10**	0.2 – 0.1 MeV	1043284	731877
9	**11**	100 – 46.5 keV	514137	648126
10	**12**	46.5 – 21.5 keV	126712	279418
11	**13**	21.5 – 10 keV	32802	103788
12	**14**	10 – 4.65 keV	2038	9687
13	**15**	4.65 – 2.15 keV	187	1179
14	**16**	2.15 – 1 keV	6	47
15	**17**	1 – 0.465 keV	0	0
16	**18**	465 – 215 eV	0	0
17	**19**	215 – 100 eV	0	0
18	**20**	100 – 46.5 eV	0	0
19	**21**	46.5 – 21.5 eV	0	0
20	**22**	21.5 – 10 eV	0	0
21	**23**	10 – 4.65 eV	0	0
22	**24**	4.65 – 2.15 eV	0	0
23	**25**	2.15 – 1.0 eV	0	0
24	**26**	1.0 – 0.465 eV	0	0
25	**27**	0.465 – 0.215 eV	0	0
26	**28**	0.215 – 0.001 eV	0	0

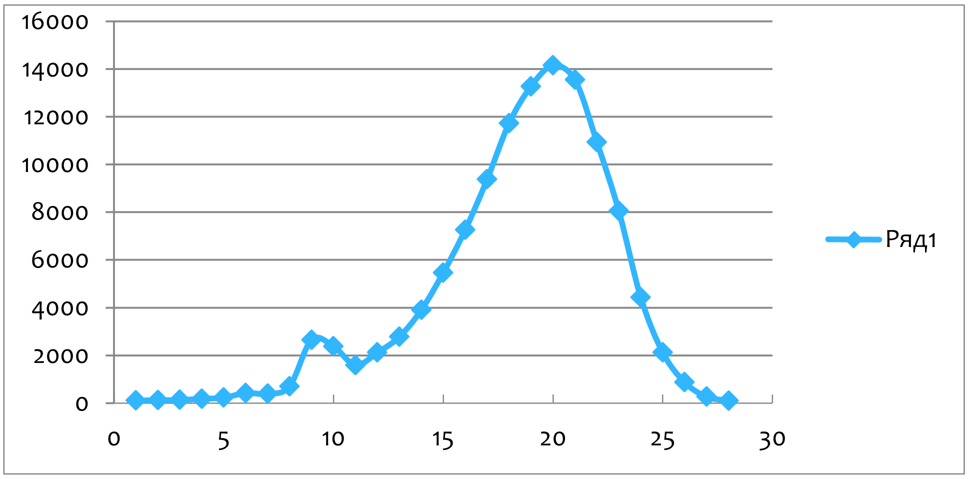

Fig.5. High density LiD sphere R=0.1 cm capture spectrum.

As we can see, spectra have maximum quantity of neutrons in neighbor energy groups and are similar to each other. Thus, capture spectrum can be recreated using outgoing spectrum which can be experimentally measured by TOF method.

Example №3 of calculated spectrum: sphere with R=0.1 cm, made of lithium deuteride LiD with density 880,000 kg/m3 which is equal to 1000 nominal densities.

It can be created by compression of nominal density LiD sphere with radius 1 cm: 10 times by radius. Isotope composition is natural: Li^6 (7.5%) and Li^7 (92.5%), 100,000 neutrons with energy 14 MeV were injected into center at t=0.

As we can note, neutron flux is trapped inside this high density assembly, besides its small radius. Only several neutrons escape from external surface of the sphere, and their spectrum strongly differs from capture spectrum.

Table 4: Calculation Li^6(7.5%)Li^7(92.5%) assembly spectra: R=0.1cm; p=880,000 kg/m^3

ABBN-78 Group №	Group № in figures	Energy interval	Outgoing spectrum	Capture spectrum
-1	**1**	14.5 – 14.0 MeV	0	108
0	**2**	14.0 – 10.5 MeV	2	124
1	**3**	10.5 – 6.5 MeV	3	134
2	**4**	6.5 – 4.0 MeV	0	181
3	**5**	4.0 – 2.5 MeV	0	227
4	**6**	2.5 – 1.4 MeV	2	423
5	**7**	1.4 – 0.8 MeV	0	390
6	**8**	0.8 – 0.4 MeV	2	705
7	**9**	0.4 – 0.2 MeV	1	2647
8	**10**	0.2 – 0.1 MeV	2	2380
9	**11**	100 – 46.5 keV	2	1594
10	**12**	46.5 – 21.5 keV	4	2118
11	**13**	21.5 – 10 keV	4	2777
12	**14**	10 – 4.65 keV	6	3902
13	**15**	4.65 – 2.15 keV	2	5463
14	**16**	2.15 – 1 keV	1	7256
15	**17**	1 – 0.465 keV	0	9376
16	**18**	465 – 215 eV	2	11729
17	**19**	215 – 100 eV	4	13275
18	**20**	100 – 46.5 eV	1	14148
19	**21**	46.5 – 21.5 eV	1	13546
20	**22**	21.5 – 10 eV	0	10941
21	**23**	10 – 4.65 eV	0	8049
22	**24**	4.65 – 2.15 eV	0	4426
23	**25**	2.15 – 1.0 eV	0	2124
24	**26**	1.0 – 0.465 eV	0	872
25	**27**	0.465 – 0.215 eV	0	267
26	**28**	0.215 – 0.001 eV	0	97

Temperature of LiD compressed sphere was chosen T=300 Kelvins.

Example №4 of calculated spectrum: metal sphere made of Tantalum-181 nominal density, R=25 cm, initial energy of neutrons 14 MeV. At t=0 were injected 100,000 neutrons into center of the sphere.

Table 5: Calculation Ta^{181} assembly spectra: R=25 cm, nominal metal density

ABBN-78 Group №	Group № in figures	Energy interval	Outgoing spectrum	Capture spectrum
-1	**1**	14.5 – 14.0 MeV	1774	196
0	**2**	14.0 – 10.5 MeV	850	19
1	**3**	10.5 – 6.5 MeV	197	0
2	**4**	6.5 – 4.0 MeV	231	11
3	**5**	4.0 – 2.5 MeV	465	148
4	**6**	2.5 – 1.4 MeV	1359	1339
5	**7**	1.4 – 0.8 MeV	3545	4822
6	**8**	0.8 – 0.4 MeV	9430	12160
7	**9**	0.4 – 0.2 MeV	15643	26135
8	**10**	0.2 – 0.1 MeV	13949	34891
9	**11**	100 – 46.5 keV	5748	35927
10	**12**	46.5 – 21.5 keV	1104	14408
11	**13**	21.5 – 10 keV	165	3255
12	**14**	10 – 4.65 keV	21	565
13	**15**	4.65 – 2.15 keV	3	151
14	**16**	2.15 – 1 keV	0	14
15	**17**	1 – 0.465 keV	0	1
16	**18**	465 – 215 eV	0	1
17	**19**	215 – 100 eV	0	0
18	**20**	100 – 46.5 eV	0	0
19	**21**	46.5 – 21.5 eV	0	0
20	**22**	21.5 – 10 eV	0	0
21	**23**	10 – 4.65 eV	0	0
22	**24**	4.65 – 2.15 eV	0	0
23	**25**	2.15 – 1.0 eV	0	0
24	**26**	1.0 – 0.465 eV	0	0
25	**27**	0.465 – 0.215 eV	0	0
26	**28**	0.215 – 0.001 eV	0	0

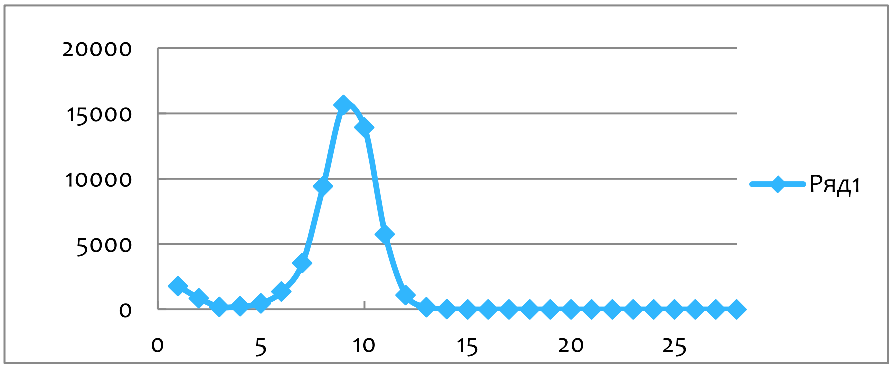

Fig.6. Metal Ta^{181} sphere R=25 cm outgoing spectrum.

During propagation from center to external surface, in this case number of neutrons due to threshold nuclear reactions grew from 100,000 to 188,527 neutrons, 134,043 of which were captured by (n,γ) reaction and 54,484 flew into vacuum. Average energy of escaped neutrons E=1.08 MeV, average diffusion time 66.4 nanoseconds. In the case of this assembly, considering its isotope composition and radius and density, both spectra are similar to each other.

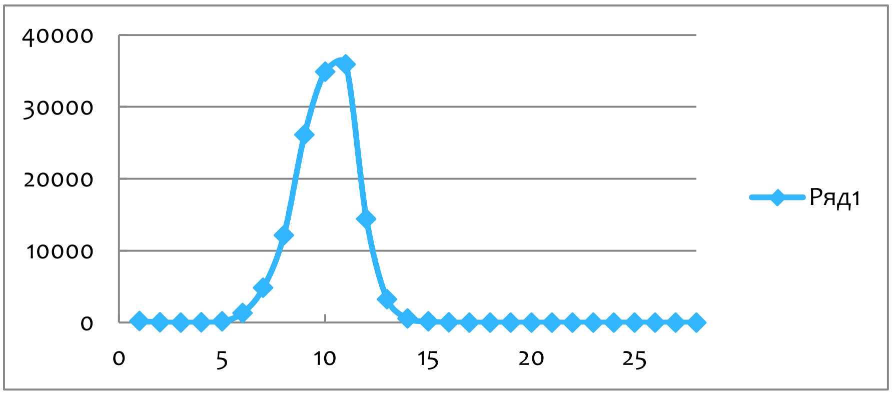

Fig.7. Metal Ta^{181} sphere R=25 cm capture spectrum.

Thus, in this case capture spectrum can be found by recalculating outgoing spectrum measured by TOF method.

It's necessary to note, that for non-fissile assemblies with nominal solid bodies densities, typical diffusion times are from several nanoseconds for radiuses several cm, to several hundred nanoseconds for radiuses around one meter when majority of neutrons, injected in the center of the sphere, are absorbed. This circumstance define required proton beam duration and TOF histogram channel duration, which are in technically achievable interval, considering both ability of electronics and ability to gather enough statistics during measurements.

Modeling results of all 4 assemblies show that leakage neutron spectrum which can be measured by TOF method, and absorption spectrum needed for calculation of fast reactor's breeding ratio, are two different neutron spectra. They even have maximum number of neutrons in different energy groups. Their correlation can be found making variant numerical calculations. In the case of big assemblies, which radius many times exceeds transport free path of neutrons, their outgoing and capture spectra have average energy around 100 keV. Such energies already can be measured by TOF spectrometer with resolution factor 6 nanoseconds/meter.

4. Abilities of the experimental equipment

In Fig.8 first 100 channels of TOF histogram, measured by fast 5-liter section of gamma-detector, are presented. Section is observed by FEU-110 photomultiplier. Data acquisition system's channel width is 100 nanoseconds, proton beam duration 250 nanoseconds. Histogram was obtained at the 50 meter TOF base of the INES TOF spectrometer, based on neutron beam of pulsed neutron source RADEX. TOF resolution factor is 6 nanoseconds per meter.

In the first 3 channels we can see prompt gamma-flash, it is successfully separated from the neutron spectrum by spectrometer without signal overloading. Its duration is equal to proton beam's duration. Histogram from He^3 counter, shown in Fig. 9, is started from the same moment of time as Fig.8, and we can see that in the first 6 channels there are no neutrons.

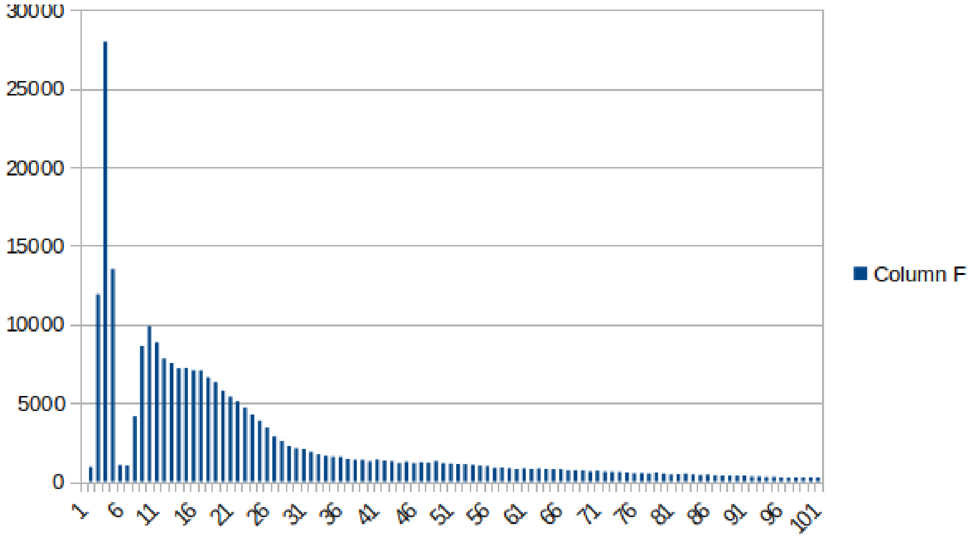

Fig.8. First 100 channels of the TOF histogram, gamma detector.

Channel number 10 corresponds to neutrons with energy 14 MeV, channel number 100 depicts neutrons with energy 140 keV. Around 10^{th} channel we can see cascade neutrons with energies up to maximum proton energy, which is 267 MeV. Such neutrons have velocity equal to 0.627 speed of light in vacuum and fly 50 meter path during 250 nanoseconds.

We can see that cascade neutrons, observable around channel number 10, are not separated on histogram from spallation neutrons which has average energy 3 MeV and energy distribution similar to fissile neutrons. Spallation neutrons continue up to 30^{th} channel, after which the Fermi spectrum (1/E) is starting.

In Fig. 9 we can see, that flash finishes only after 50^{th} channel that depicts that He^3-counter has average response time 2 microseconds.

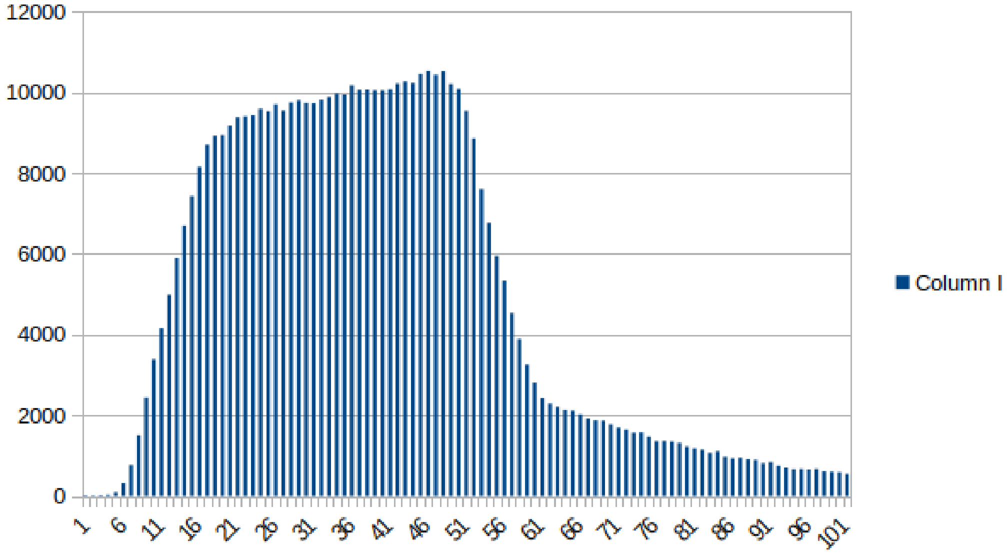

Fig.9. First 100 channels of the TOF histogram, He^3 counter.

Resonance filter aluminum Al^{27} total cross section resonance around 140 keV is exactly observed on histogram of the gamma detectors. Also are observed total cross section Al^{27} resonance at 35 keV and Fe^{56} resonance around 28 keV.

On He^3 counter's histogram, delta tau in TOF resolution formulae corresponds to 2 microsecond of He^3 counter response time, instead of 250 nanoseconds beam. Due to less energy resolution, 140 keV aluminum resonance is not resolved, while 35 keV Al^{27} and 28 keV Fe^{56} resonances interfere each other and are observed as single resonance.

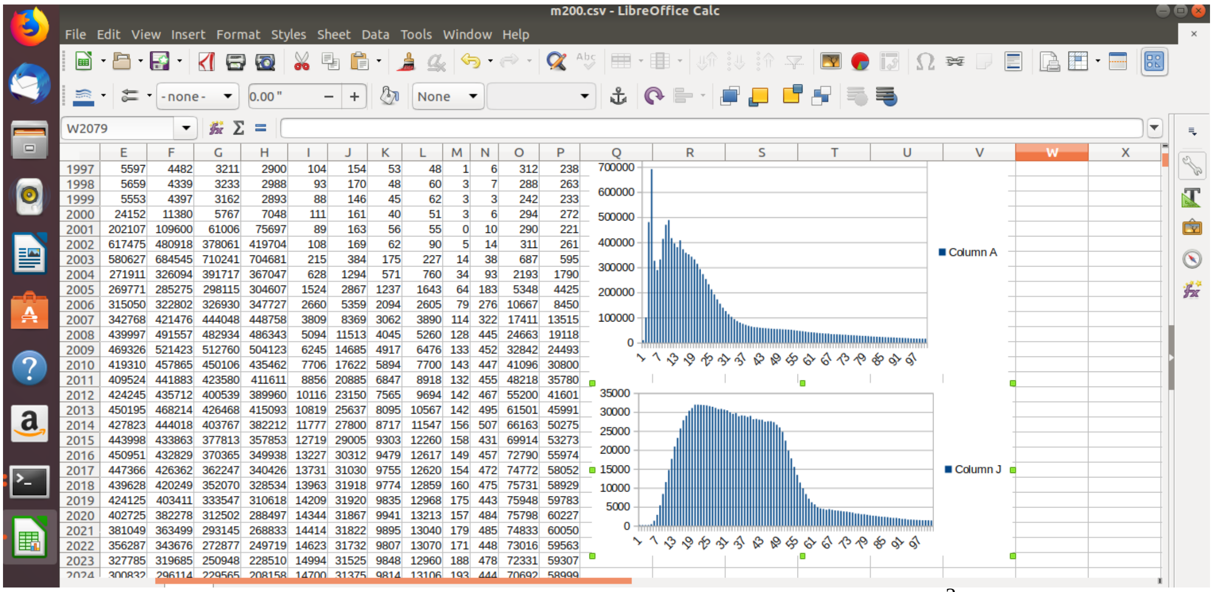

Fig.10. Typical view for 16 channels data, 8 gamma-detectors and 8 He^3-counters, high energies.

Thus, we can see that the TOF spectrometer 'INES' allows to measure histogram and resolve wide cross section resonances on gamma detectors up to energies around 140 keV where are situated majority of neutrons in the cores of fast breeder reactors with diluted fissile material (where majority of neutrons in modeling Figs. 2−7 are situated on energy axis).

TOF resolution 6 ns/meter, achieved at 'INES' TOF INR RAS spectrometer, is enough to solve tasks for fast neutron spectrometry, resolving spectra from subcritical assemblies. Comparing experimentally measured spectra with calculated predictions, it's possible to check them, to modify calculation models and to customize neutron group constants.

5. Conclusion

Neutron spectra of large fast breeder reactors with diluted fissile material have average energy around ~150 keV. This value turns out measurable already by TOF spectrometers, which have resolution factor 6 nanoseconds per meter and better.

Outgoing spectrum and capture spectrum differ one from another. Outgoing spectrum can be measured by TOF method, while capture spectrum is needed to calculate fast neutron reactor's breeding ratio. Thus, using experimentally observable TOF spectrum, it's necessary to reconstruct capture spectrum using numerical modeling.

Due to activation of experimental assembly, average proton beam power is limited by value around 1 kW, for subcritical assemblies with easily changeable material composition.

It's necessary to note, that such beam power in the mode of short proton pulses, can be provided by proton linac, even without proton beam storage ring.

Helium-3 counters are traditionally used for total cross sections measurements. However, their average response time delay is around 2 microseconds and differs in large interval, depending on distance between scintillation point inside detector and central wire electrode. Thus, He-3 counters can be used only as accelerator's intensity monitor, during measurements of spectrum's upper energy part.

Measurement of subcritical assemblies' neutron spectra in 28 energy groups of ABBN-78, with TOF resolution factor 6 ns/meter, is possible. Possibility to accumulate enough statistics per energy group we also proved experimentally. At the same time, for leaking neutron flux spectra measurements in 299 energy groups of ABBN-93 system, better energy resolution factor is required.

INR RAS linear proton accelerator plans to increase proton energy up to 423 MeV, pulsed proton current up to 16 mA and achieve minimal proton beam duration 50 nanoseconds, which is realistic compared to existing mode 250 nanoseconds. Value of 50 ns beam duration with 50 nanoseconds step of data acquisition system's histogram, is effective also due to comparable value of neutron diffusion time of existing tungsten-water proton beam target at neutron spectrum energies around 100 keV, where neutron spectrum maximum of big assemblies is situated.

References

1. Stavissky Yu.Ya. Neutron Research based on Moscow Meson Factory. In: Program of experimental research at Meson Factory INR RAS, volume 4, Moscow, 1986, p.7−21.
2. Grigoriev Yu.V. Dissertation, p.62−67. Obninsk, 1980.
3. A.V. Dementyev, N.M. Sobolevsky. SHIELD – Universal Monte Carlo hadron transport code: scope and application. Radiation Measurement 30 (1999) p.554−557.
4. https://www.inr.ru/shield
5. https://www.bnl.gov
6. L.P. Abagian, N.O. Baziants, M.N. Nikolaev, A.M. Tsybulia. Gruppovye konstanty dlia rascheta reactorov & zashity. Handbook. Moscow, Energoatomizdat, 1981 (in Russian).