TOF METHOD MEASUREMENTS AT INR SPALLATION NEUTRON SOURCE RADEX

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Abstract. Installation INES based on pulsed spallation neutron source RADEX has been developed at INR RAS, Moscow, Russia. INES is designed for investigations of neutron cross sections for reactor fuel and construction material alloys. INES uses 50 meter spectrometer base, which together with 500 ns proton beam duration of accelerator makes possible to measure group total and group capture cross sections of researched materials. Cross sections are experimentally observed and measured with corresponding block-effect and Doppler-effect values, with automatic averaging of resonance structure of all isotopes, available in alloy. Measured results are used for calibration of calculation codes, which are used in the process of design and certification of nuclear reactor cores and their biological radiation protection shields.

Installation INES uses four Helium-3 detectors as beam intensity monitors, four Helium-3 detectors as transmittance functions detectors and 8-sectional liquid (n,γ) detector which scintillator's volume is 40 liters.

During year 2017 new fast 16-channel data acquisition system was developed, tested and employed. It uses modern electronic element base, has 100 nanosecond time step in 16-channel mode and 50 nanosecond step in 8-channel mode. This data acquisition system is designed to work in multiplicity mode, which provides good separation between effect and background during capture (n,γ) and fission (n,f) cross sections measurements. At beam frequency 50 Hz, statistics is being collected into 200000 channel histogram. Exact spectrum and cross sections measurements will enable more exact calculations of fast breeder reactor cores characteristics, including critical mass and breeding ratio.

1. Introduction

One of the most important tasks, for which INR RAS proton linac was designed and built, were TOF measurements to provide fast neutron reactors program by exact neutron group cross sections data. Powerful linac with one-turn beam extraction storage ring for high intensity, and long TOF bases for high $\left(\frac{\Delta t}{L}\right)$ parameter were designed. Target reloading machine allows to change proton beam target making modeling of the reactor's core and measure corresponding neutron spectrum.

Project parameters of the proton linac of INR Russian Academy of Sciences are: proton energy 600 MeV, pulsed current 50 mA, frequency 100 Hz, maximum pulse duration 100 (200) mks; They provide average current 0.5 (1.0) mA, mean beam power 300 (600) kW, neutron yield into angle $4\pi \sim 10^{17}$ neutrons per sec.

Corresponding to the technical project, linac is equipped by the storage ring which has project parameters: beam aperture 200 mm, circle length 2πR = 102.8 meters, proton cycling period 430 nanoseconds at 600 MeV. Maximum pulse current, allowed by space charge is 10 Amperes at 600 MeV, it corresponds to maximum pulse intensity $2.3 \cdot 10^{13}$ protons in each pulse at 100 Hz and average neutron flux with W target in short pulses at 100 Hz, up to

$2.5 \cdot 10^{16}$ n/s. Storage ring now is under construction. Significant part of its radiation sustainable equipment is already manufactured.

2. Accuracy requirements

Requirements for accuracy of group neutron cross sections for many types of reactors are determined by share of delayed neutrons of employed fissile nuclide and shown in table 1.

Table 1

Reactor fuel composition	U^{235}	U^{233}	Pu^{239}	BN-1200 nitride fuel
Desirable accuracy of criticality coefficient calculation (ΔK/K), %	0.6	0.2	0.2	0.42

Diffusion one-group approximation gives expression for critical radius

$$R_{crit} = \frac{\pi}{\sqrt{3(K_{inf}-1)\Sigma_a \Sigma_{tr}}} - \frac{0.71}{\Sigma_{tr}} \quad (1)$$

And expression for critical mass

$$M_{crit} \sim R_{crit}^3 \sim (\Sigma_a \Sigma_{tr})^{\frac{3}{2}} \sim (\sigma_a \sigma_s)^{\frac{3}{2}} \quad (2)$$

Accuracy of neutron cross sections must provide exactness of critical mass prediction $\left|\frac{(M_{calc}-M_{measured})}{M_{measured}}\right|$ better then value, which corresponds to maximum allowed $\left(\frac{\Delta K}{K}\right)$ value.

Accuracy of TOF transmittance method for measurements of total cross sections for one channel of data acquisition system is defined by expressions:

$$N(x) = N_0 \exp(-nx\sigma_{total}),$$
$$T = \frac{N_x}{N_0},$$
$$\sigma_{total} = \left(\frac{1}{nx}\right) ln\left(\frac{1}{T}\right),$$
$$\frac{(\delta\sigma_{total})}{(\sigma_{total})} = \left(\frac{1}{\sqrt{N_0}}\right)\left(\sqrt{\frac{1+T}{T}}\right)\left(\frac{1}{ln\left(\frac{1}{T}\right)}\right). \quad (3)$$

For total cross section accuracy 0.2% it's necessary to accumulate statistics ~1000000 counts in each histogram channel. Coefficient which depends on pattern thickness is shown on fig.1.

Achieved accuracy of fast breeder reactors criticality predictions $\frac{\Delta K}{K}$ is shown in table 2.

Table 2. Accuracy of existing nuclear data and their calculation codes

Neutron constant system	MCNP	ABBN-78	ABBN-93
$\frac{\Delta K}{K}$, %	2	2	0.5

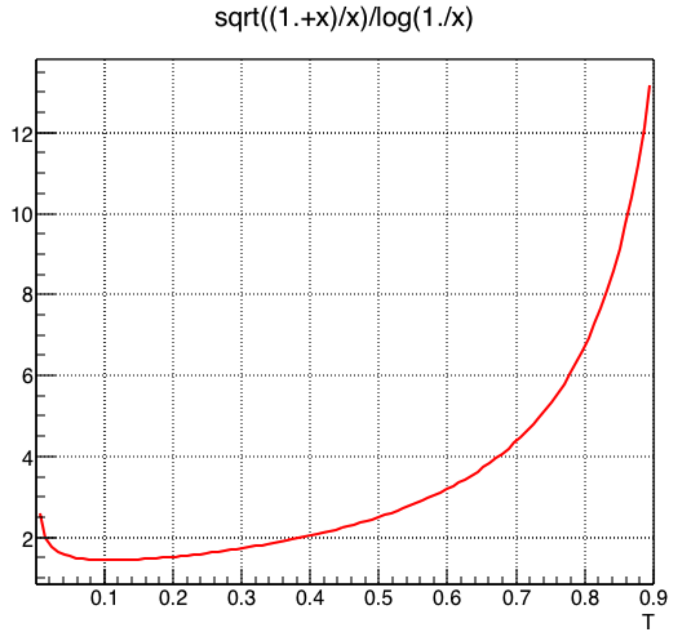

Fig.1: Coefficient proportional to pattern thickness.

During fact neutron moderation, number of scatterings between energies E_0 and E is

$$N_{scatterings} = \left(\frac{1}{\xi}\right)\ln\left(\frac{E_0}{E}\right), \qquad (4)$$

where $\left(\frac{1}{\xi}\right) \approx \frac{A}{2} + \frac{1}{3} + \frac{1}{18A}$.

If E_0=14 MeV and E=0.0253 eV, for substance like Li_6D we find that 28 energy groups in ABBN-78 are enough.

Due to presence in the core of fast breeder reactor such medium atomic mass nuclides like Na, Fe, Cr, Ni, Ti, in neutron group cross section constants ABBN-93 were chosen 299 groups. In our measurements we are working to fit ABBN-93 requirements.

Energy resolution of TOF spectrometer as function of $(\Delta t/L)$ parameter is shown in table 3. Here, $\Delta E=2.77\cdot 10^{-5}\cdot E^{3/2}\cdot(\Delta t/L)$, Δt – nanoseconds, L – meters, E – eV.

Table 3

$\Delta t,\ \left(\frac{ns}{m}\right)$	$\Delta E,\ eV$						
	E=10 eV	E=100 eV	E=1 KeV	E=10 KeV	E=100 KeV	E=1 MeV	E=10 MeV
100	0.09	2.8	88	2800	88000	2800000	88E6
500	0.04	1.4	44	1400	44000	1400000	44E6
10	0.009	0.28	8.8	280	8800	280000	8.8E6
5	**0.004**	**0.14**	**4.4**	**140**	**4400**	**140000**	**4.4E6**
1	0.0009	0.028	0.88	28	880	28000	880000
0.5	0.0004	0.014	0.44	14	440	14000	440000
0.1	0.00009	0.028	0.088	2.8	88	2800	88000

Value 5 ns/m, which is marked in table 3 by thick line, corresponds to our measurements on installation INES.

Influence of Doppler-effect and cross sections self-shielding of multi-isotope compounds on reactor criticality and other parameters on example of one-group diffusion theory

$$D\Delta\Phi - \Sigma_a\Phi + \upsilon\, \Sigma_f\Phi = 0\,, \qquad (5)$$

$$D\Delta\Phi + (\eta - 1)\, \Sigma_a\Phi = 0, \qquad (6)$$

thus we get differential equation with Laplasian and buckling parameter

$$\Delta\Phi + (\eta - 1)\left(\frac{\Sigma_a}{D}\right)\Phi = 0 \qquad (7)$$

or the same equation in ordinary form

$$\Delta\Phi + \left(\frac{\eta-1}{L^2+\tau}\right)\Phi = 0, \qquad (4)$$

where D – diffusion coefficient, Φ – neutron flux, Σ_a – macroscopic absorption cross section, τ – Fermi's age of neutron flux, L^2 – square of diffusion's length.

Lower resonance levels have mainly neutron capture properties. Doppler-effect and internal block-effect influence on value of Σ_a and are measured by gamma-detectors. Upper resonance levels are mainly scattering neutrons without absorption them. At the same time Doppler-effect and multi-isotope alloy's interference of resonance levels influence on value D. Thus, leakage from fast neutron reactor's core into reflector also changes at different temperatures and dilutions. This effect if researched by transmission functions measurements in TOF experiments. Both first and second effects are able to change effective L^2 and τ.

In TOF experiments transmission and self-indication functions must be measured in unresolved resonance area that is possible using multiplicity coincidence method.

It's necessary to mention that into one energy group of ABBN-93 many histogram channels are coming, so as energy width of one group grows in high energy region as shown in table 4.

Compared to resonance structure measurements, where energy level's width Γ_γ is approximately constant in wide energy range and is about 0.1 eV for heavy nucleus, group cross sections measurements allow to measure up to higher upper energy level using the same TOF spectrometer with the same energy resolution ($\Delta t/L$) measured in nanoseconds per meter. It is also important that number of counts per channel, in expression for accuracy of measurements, corresponds not to single data acquisition system channel, but to energy group where present many histogram channels.

Table 4. Group energy width, eV, of the ABBN-78 neutron constants

	Energy width ΔE, eV of group number N						
	E=10 eV	E=100 eV	E=1 KeV	E=10 KeV	E=100 KeV	E=1 MeV	E=10 MeV
Group's number	21	18	15	12	9	6	1
Width	11.5	115	1150	11500	115000	600000	3500000

At the same time, capture cross sections measurements must be done in the energy area of unresolved resonances using multiplicity coincidence method, which gives an opportunity to distinguish effect and background.

3. Experimental installation INES

Installation INES is shown on fig.2.

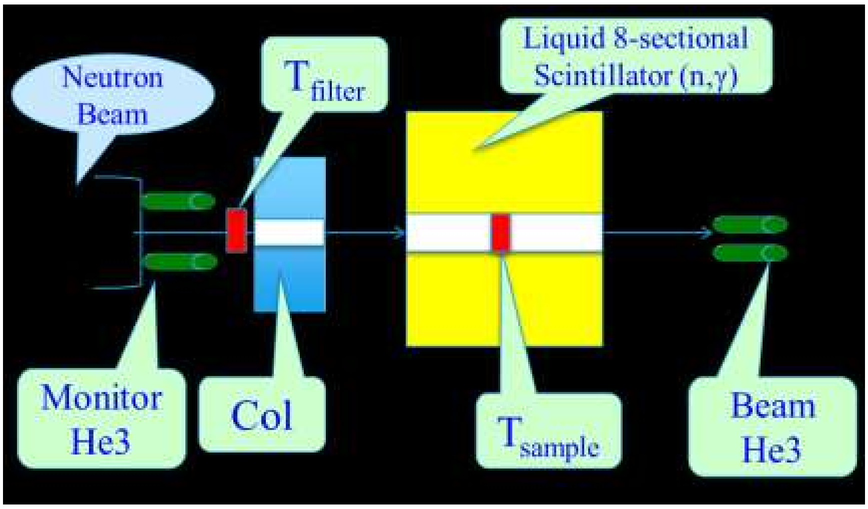

Fig. 2: Installation INES.

Corresponding to fig. 2, neutron beam arrives from the left side in the evacuated neutron guide. Four monitor He-3 counters are in beam. After that, patterns-filters like Mn-55 are installed, and beam is collimated for liquid scintillator detector's aperture. Measured isotope plate is installed to the center of the scintillator tank. From the right side, before beam capture, four He-3 counters are installed to measure total cross section.

During the described our measurements, pure patterns of Gold Au-197 and Tantalum Ta-181 were used. In figs.3 and 4 we can see the best world data taken from BNL. For Ta-181 we can see resolved resonances area up to neutron energy 2500 eV.

In the figure 5 our INR measurements of Ta-181 are shown. On 'X'-axis is histogram channel number, on 'Y'-axis number of counts.

During measurements of Au-197 and Ta-181, proton linac was working with parameters: proton energy 209 MeV, pulsed current 10 mA, pulse width 1 mks, frequency 50 Hz. Installation INES was installed on 50 meter TOF base of spallation pulsed neutron source RADEX.

In fig. 6, work with experimental histogram is shown. Experimental spectral histograms can be transformed into cross section curves by main two methods. Below few hundreds eV for heavy atoms with high resonance level densities, where ΔE of spectrometer resolution function is smaller than Γ_γ which is about 0.1 eV, the method of form can be used. At higher neutron energies in the resolved area it's necessary to use method of square areas.

Program for resonance parameters definition automatically calculates: background level as function of neutrons energy on experimental histogram like fig.7, energy of the center of resonance area, width of resonance on half-altitude, area of the resonance.

In fig. 8 we can see action of Mn-55 filner: in its 336 eV resonance area, all counts are background. Observing this effect makes possible to calculate the background component as energy's function and extract it from the spectra.

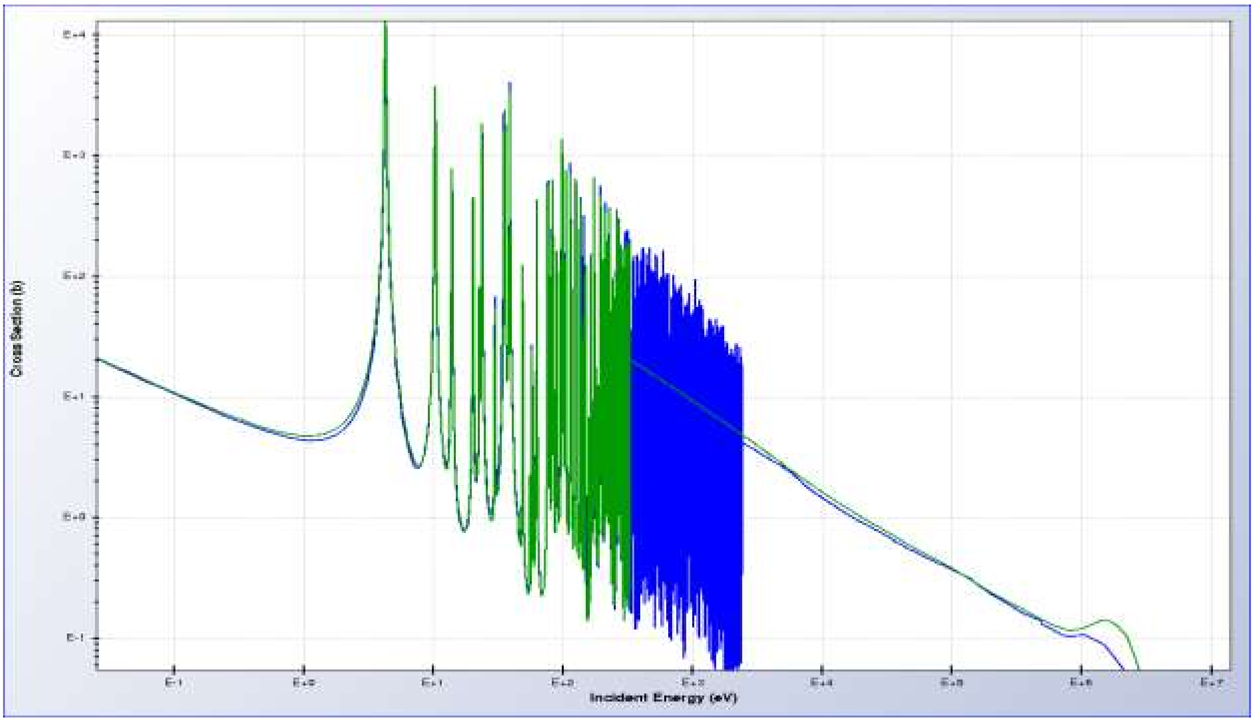

Fig. 3: The best world data for Ta-181 capture cross section in wide energy region.

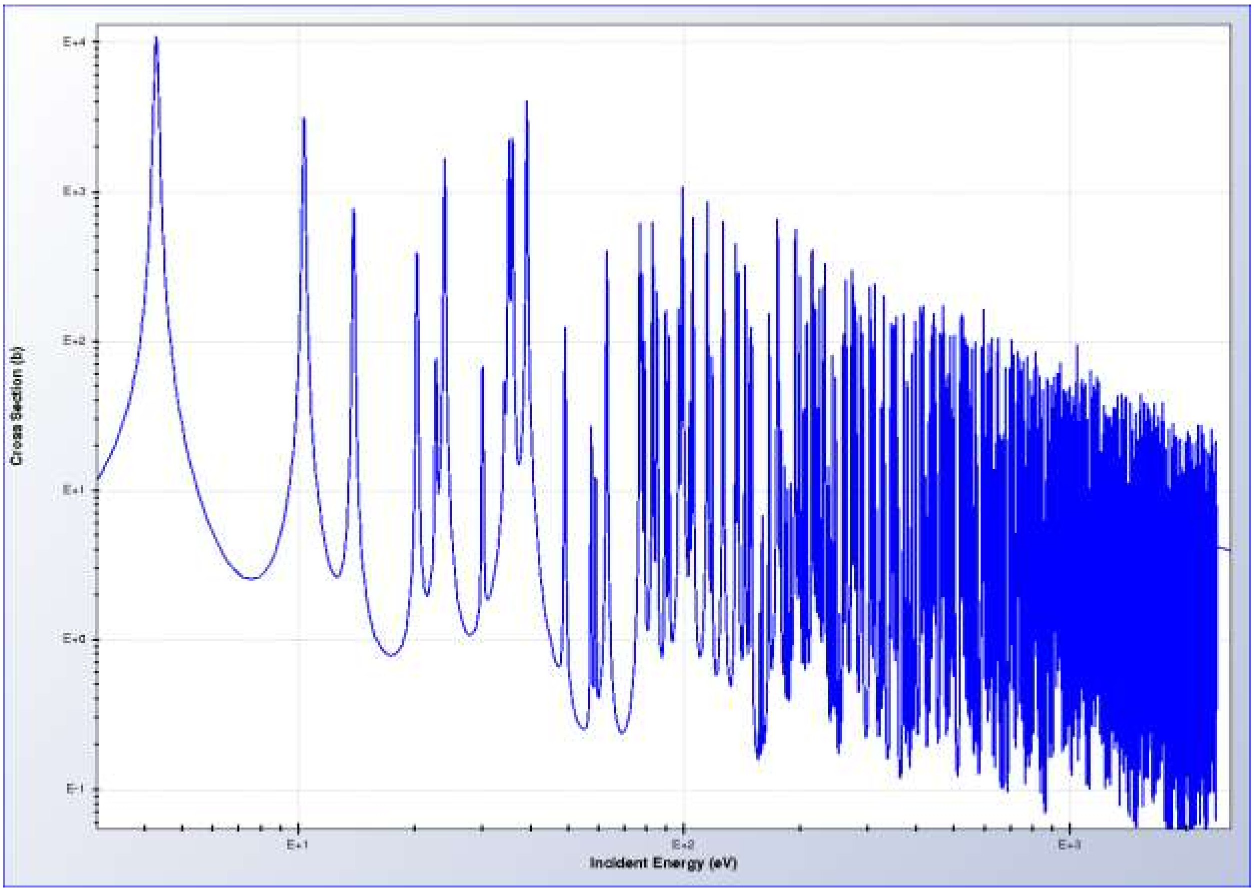

Fig. 4: Ta-181 capture cross section area of the resolved resonances, up to 2500 eV.

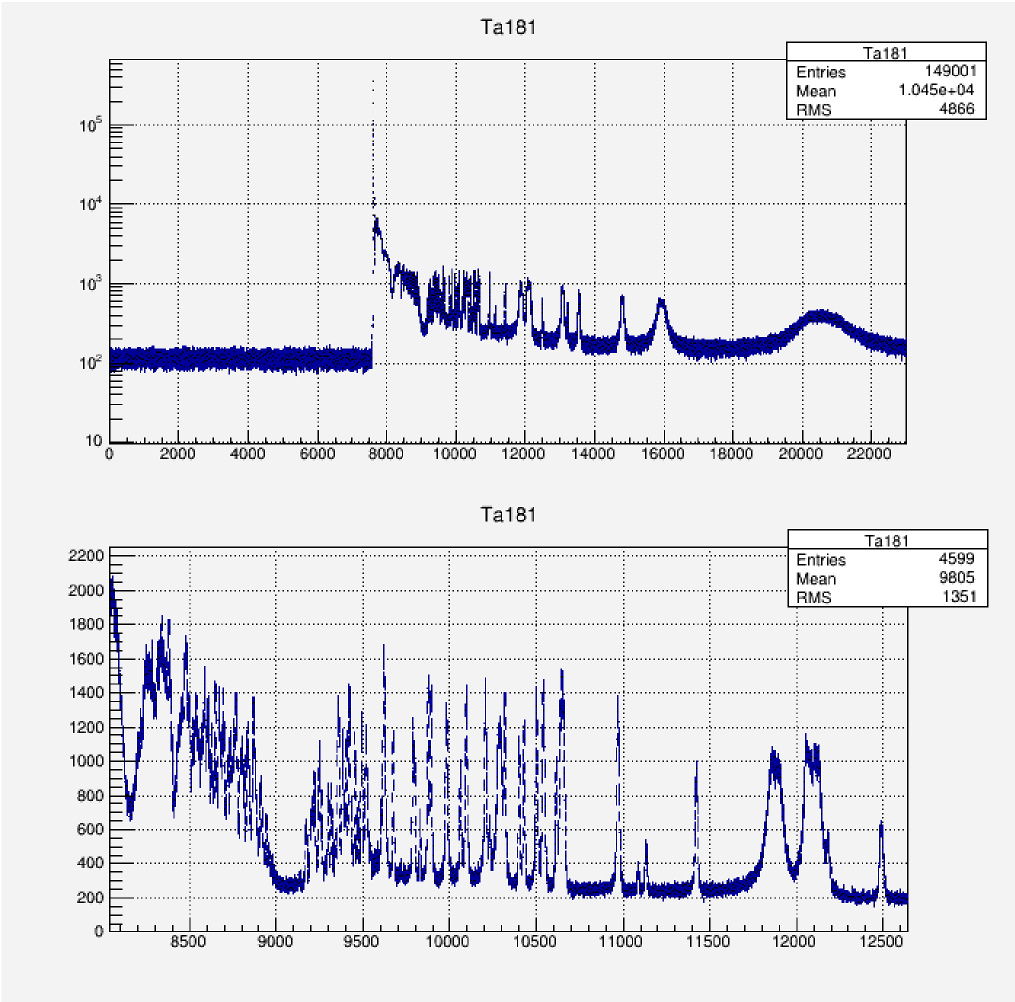

Fig. 5: Ta-181 capture cross section.

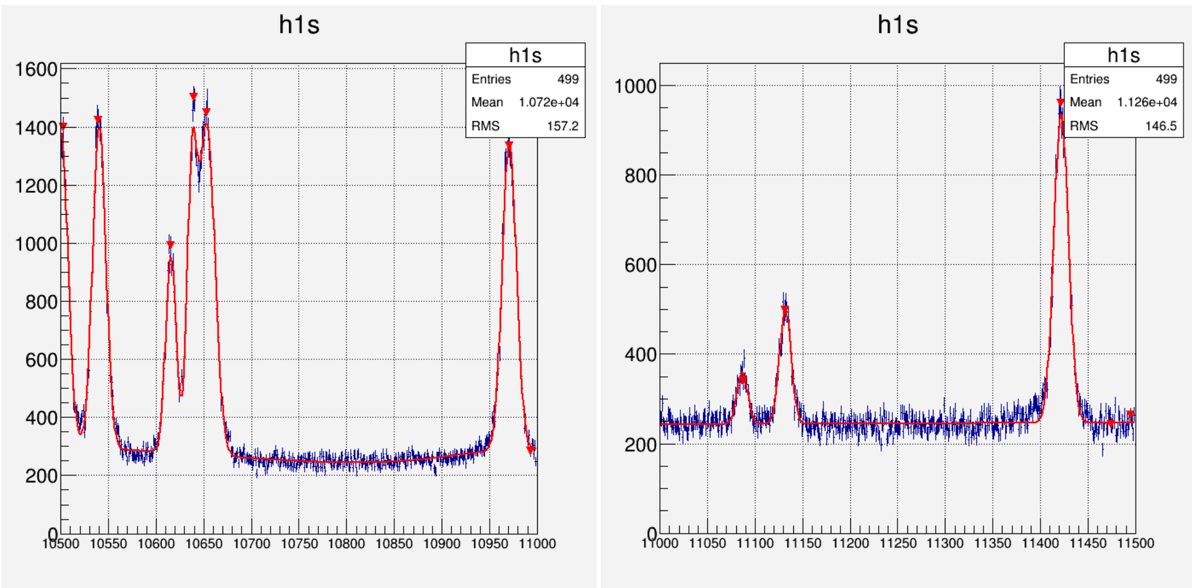

Fig. 6: Separation of effect from background.

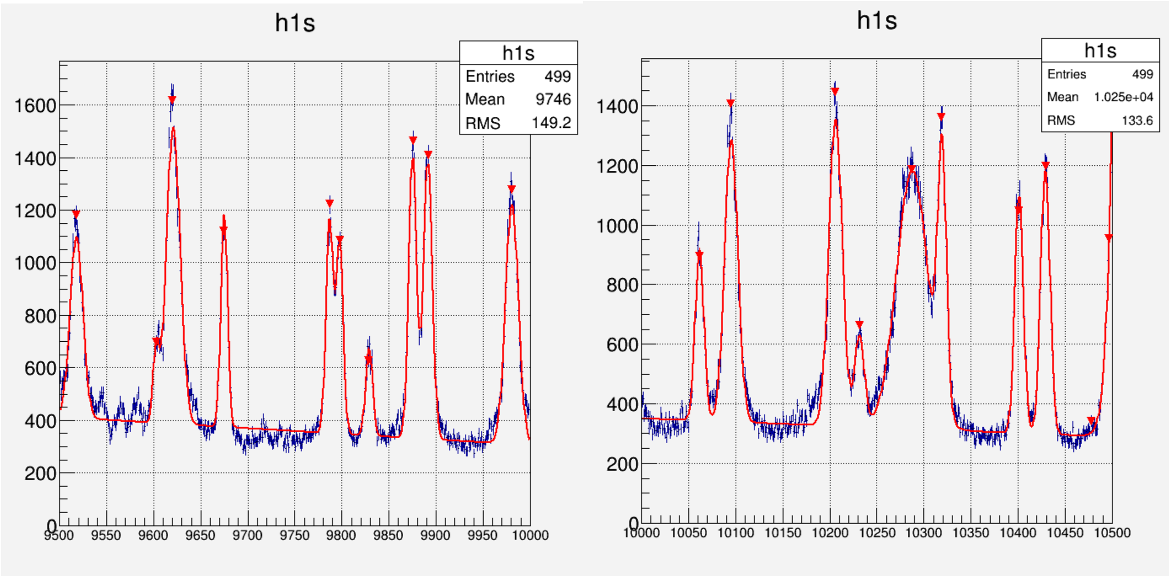

Fig. 7: Ta-181 capture experimental spectra.

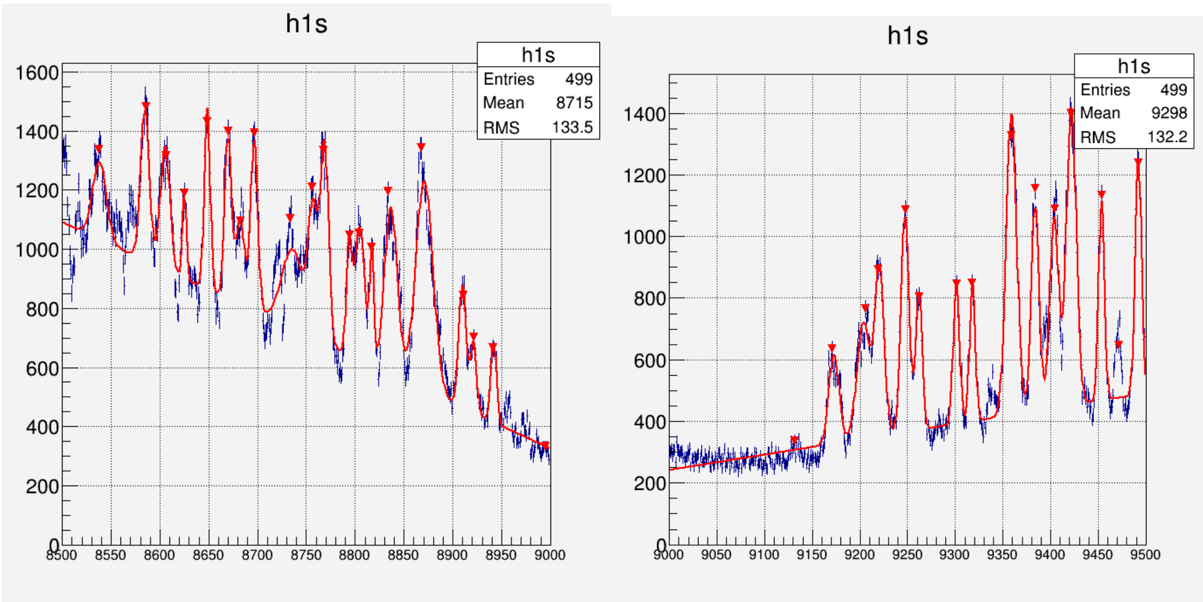

Fig. 8: Ta-181 spectrum between channels number 8500 and 9500.

At energies upper then few hundreds eV, in the case of heavy nuclides with most dense energy level system and the smallest intervals between resonances, spectrometer resolution function's value is much larger then energy width Γ_γ with doppler-effect. Due to this, experimental histogram gives us Gauss resonance parameters. When we know them, using another program for each resonance we transform the curve into the cross section curve descripted by Breit-Wigner parameters.

Unresolved area is illustrated by fig. 9. Also it's necessary to mention, that researched pattern inside the detector becomes radioactive during measurements.

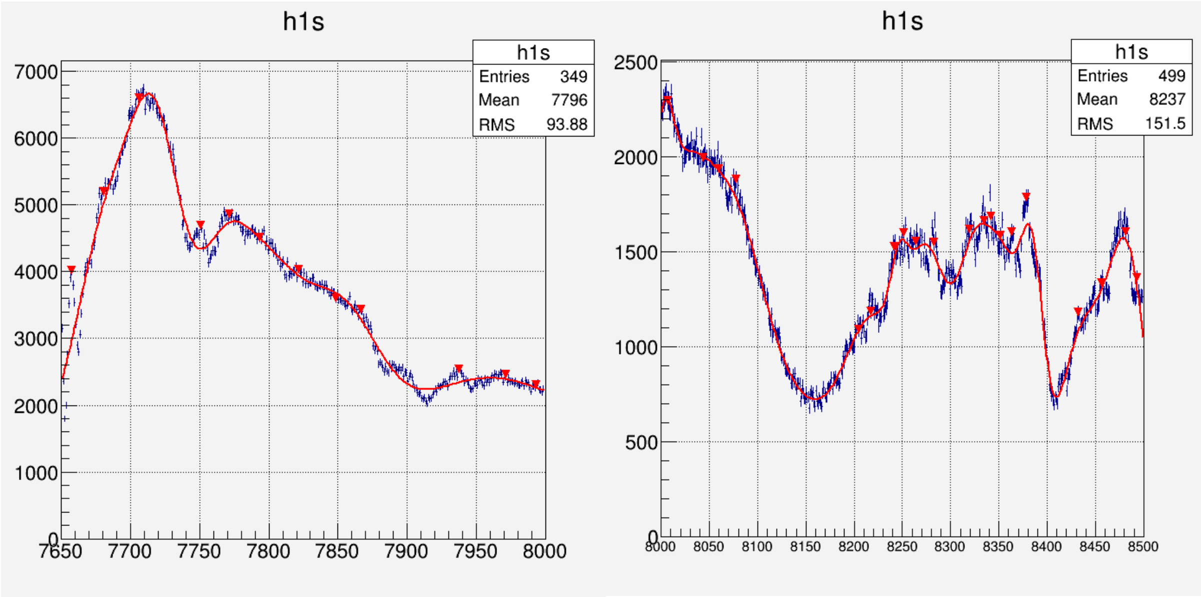

Fig. 9: Ta-181 unresolved capture resonance area after the proton beam flash.

Isotope	D_0, eV	Isotope (A+1)	T(1/2) of (A+1)
Ta-181	1.13	Ta-182m2	15.8 minutes
In-115	1.9	In-116m1	54 minutes
Au-197	1.15	Au-198	2.7 days

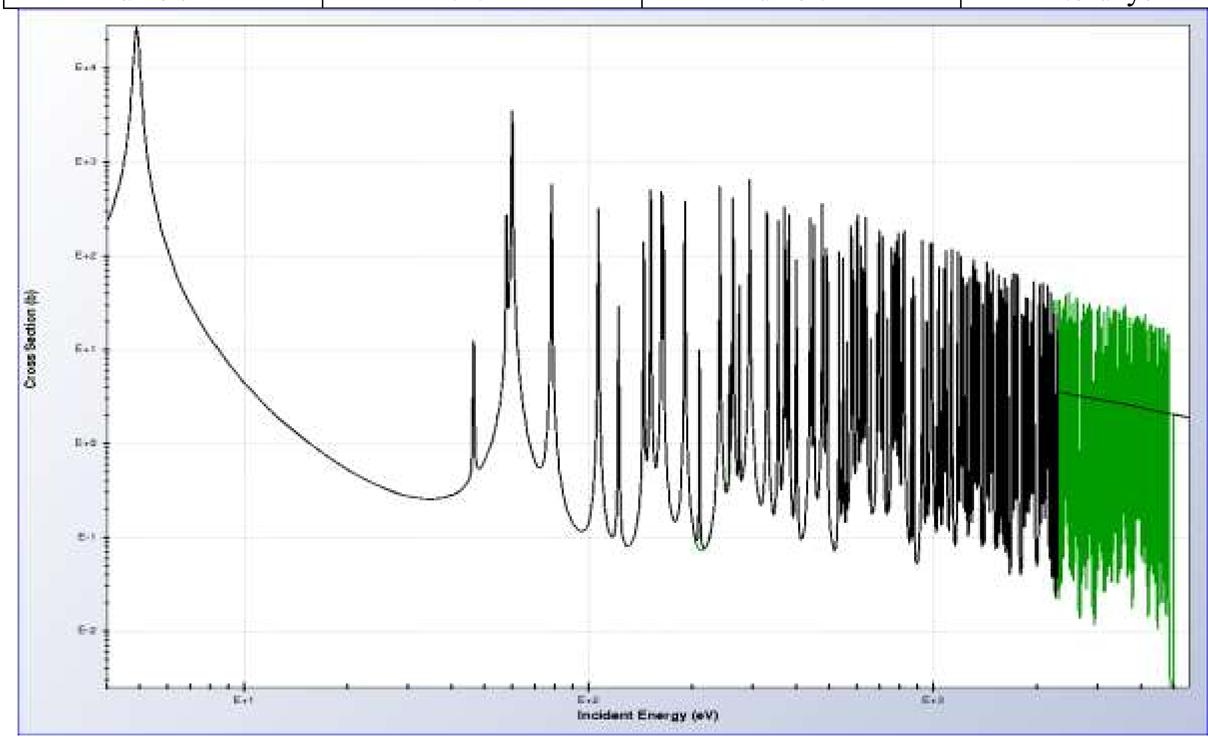

Fig. 10. Capture cross section of Au-197.

In fig.10 we can see, that in $Au^{197}(n,\gamma)$ cross section, up to 5000 eV resonances are resolved in BNL data base.

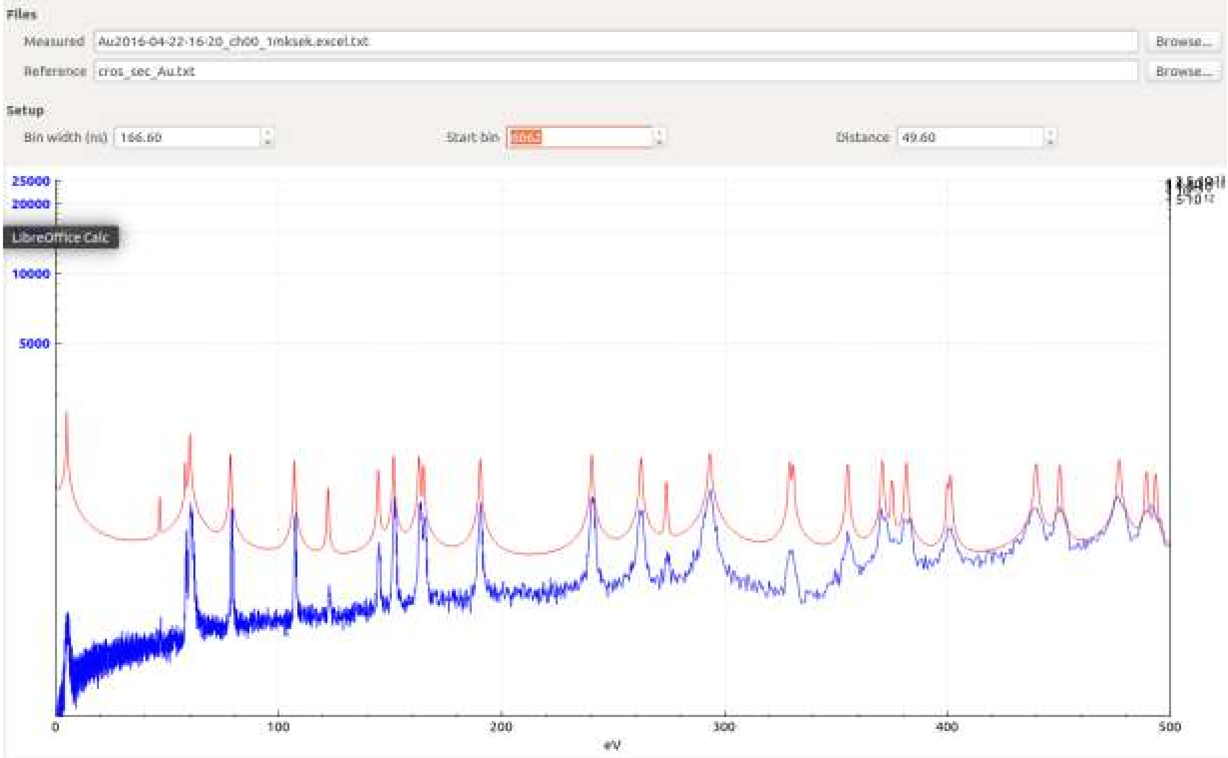

Fig. 11: Au-197 cross capture, beam pulse duration 1 mks.

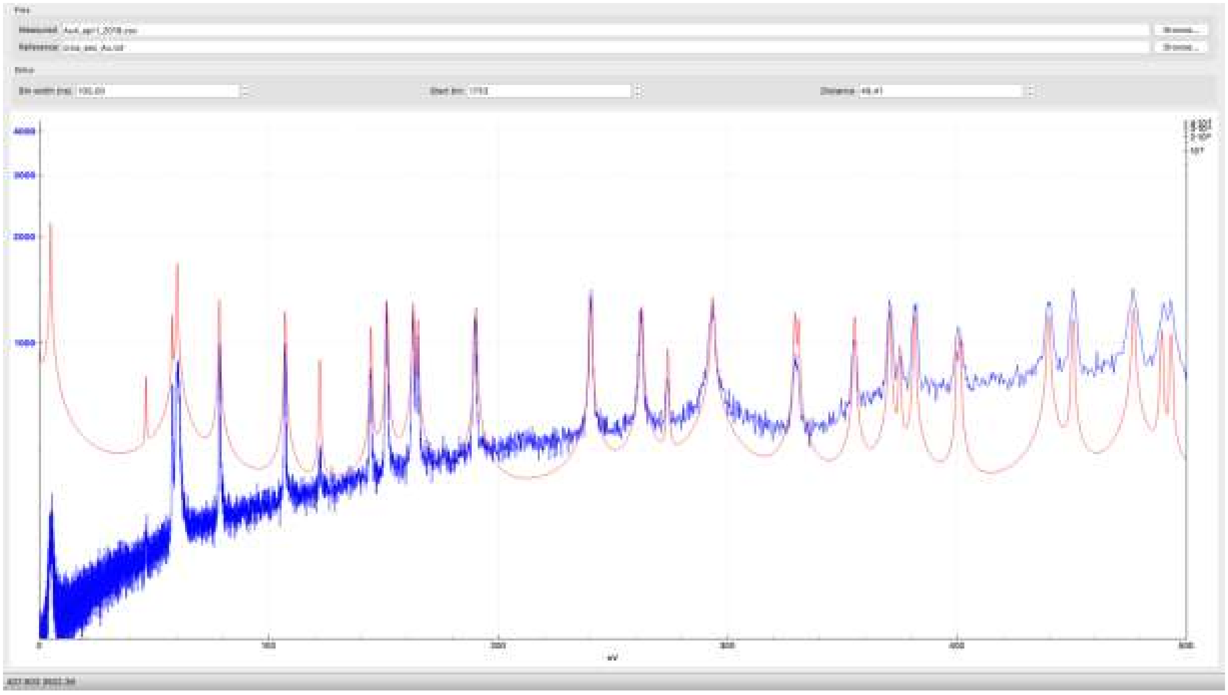

Fig. 12: Au-197 capture cross section, beam pulse duration 500 nanoseconds.

In figs. 11 and 12 we can see, that measurements at 50 meter base with proton beam 500 nanoseconds give much better results compared to 1 mks beam.

4. New equipment

During years 2017 and 2018, in our group new data acquisition system (see fig. 13) was designed, manufactured, tested and used in measurements. It has two modes: 16 signal channels at 100 ns histogram channel width and 8 signal channels at 50 ns histogram channel width. Number of histogram channels 200000 in the first and 400000 in the second mode is optimized for spectra measurements at 50 Hz. Multiplicity measurements can be made in both modes.

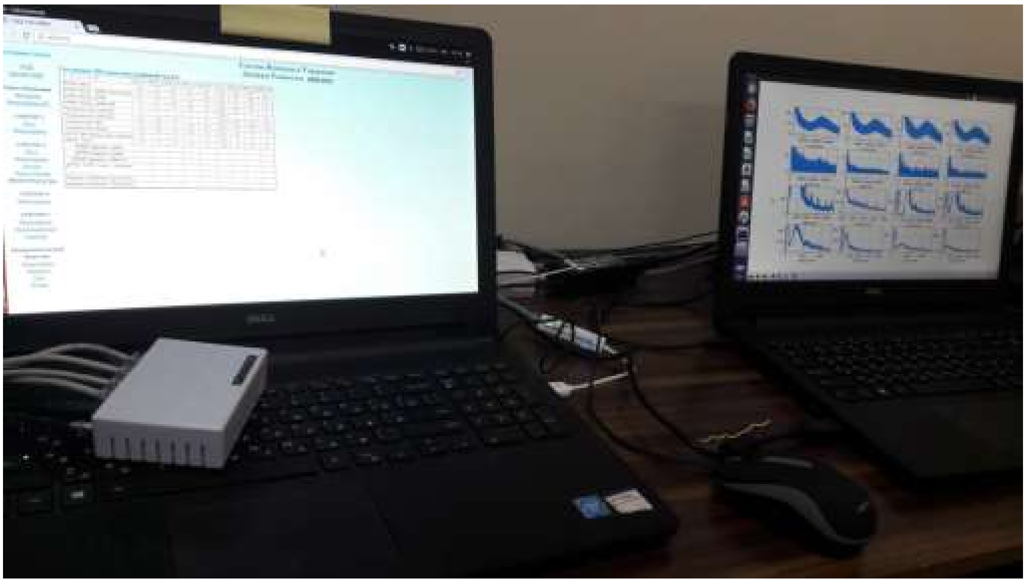

Fig. 13: New data acquisition system.

Software for new 16 channel multiplicity data acquisition system is also written in our group using PYTHON. Program allows to interactively observe data on all 16 channels in detail.

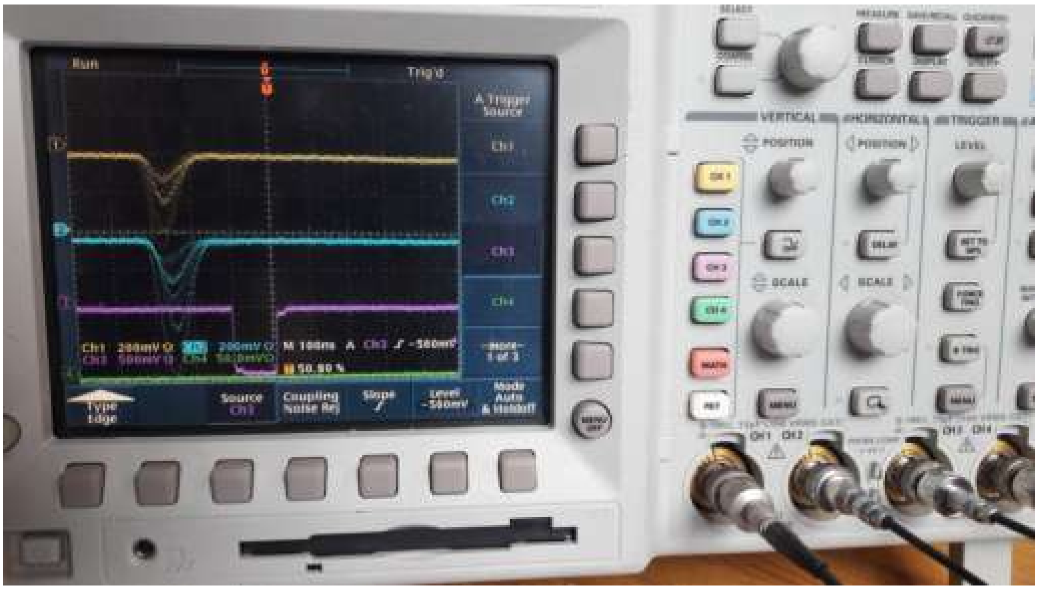

Fig. 14: testing of scintillator signals using Co-60 gamma ray source.

In fig. 14 scintillator signals after pre-amplifier are shown. They are around 60 nanoseconds at half-altitude.

New 16-channel multiplicity data acquisition system was tested at first using Co-60 gamma source and Cf-252 spontaneous fission neutron source, than it was tested on pulsed beam.

During turning, high sustainability to radio frequency transients was achieved. After turning and testing, 16-channel multiplicity system was used in measurements at TOF base 50 meters during accelerator's work with proton bean duration 500 ns.

TOF histograms of Ta-181, In-115, and Au-197 were successfully measured using 40 liter liquid scintillator gamma detector, four He-3 counters SNM-18 as neutron beam monitors and four He-3 neutron counters SNM-17 as transmission detectors.

5. Conclusion

Installation INES has properties, high enough for measurements of total and capture cross sections of reactor and construction materials in energy groups of ABBN-78 and ABBN-93 neutron constants, which are used for core criticality calculations during construction of fast breeder reactors BN-800 and BN-1200.

Internal block-effects for reactor alloys and radiation shield materials resonance cross section structure, also as Doppler-effect, can be measured. Measurements can be done both for separated isotopes of natural multi-isotope mixtures, and for multi-isotope mixtures of many chemical elements like stainless steel.

During the nearest future we plan to measure new reactor alloys group cross sections, including burning absorber reactor materials for isotope separation quality control. Also measurements of total and capture cross sections of separated isotopes, measurements at different thicknesses and temperatures in ABBN-93 energy group intervals.

Proton linac shows good progress. Operation with proton energy up to 423 MeV compared with increase of linac pulse proton current to 16 mA is planned for 2019 nearest years.

TOF measurements with 300 nanosecond proton beam are planned for 2019. Also frequency 100 Hz achievement is in progress. TOF stations at 250 meters and at 500 meters are discussed. Proton storage ring is under construction.

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