CAPTURE NEUTRON CROSS SECTIONS MEASUREMENTS OF RARE EARTH ISOTOPES

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Results of TOF measurements for total and capture neutron cross sections on isotope Ho^{165}, conducted on pulsed spallation neutron source RADEX, are presented. Pulse duration of accelerator's proton beam 250 nanoseconds, combined with 100 nanoseconds steps of data acquisition system and 80 nanosecond pulses of (n,γ) detector provided, at 50 meter base of vacuum neutron guide, value of TOF spectrometer resolution 6 nanoseconds per meter.

Measurements were done during linear proton accelerator's work for our task in years 2020 and 2021. Beam parameters were: proton energy 247 and 305 MeV, pulsed proton current 0.01 Amperes, pulse duration 250 nanoseconds, frequency 50 Hz. Average beam power 40 W provided average neutron intensity on Ho^{165} pattern 4000 n/(cm^2sec). Recycle neutrons were cut using cadmium filter.

Experimental results were compared with 4 world neutron databases: ENDF /B-VII.1, JEFF-3.1, JENDL-4.0, and ROSFOND. New experimental data on cross section resonance structure of Ho^{165} were achieved and are presented.

1. Introduction and description of the task

Neutron cross section measurements are necessary both for fundamental and applied nuclear physics. For fundamental purposes it's necessary to investigate detailed resonance structure and resonance parameters in the neutron energy region of resolved resonances, for each partial type of cross section: energy of each resonance, cross section at maximum point, it's gamma and neutron energy width, spin, average energy distance between resonances. This requires high energy resolution of TOF measurements.

For applied purposes the task is to measure partial components of cross section for each isotope in wide energy groups: both in resolved energy area and also in the high energy area of unresolved resonances.

Group cross sections, like ABBN-78, are used as initial data for calculation codes during creating the core of nuclear reactors of different types and radiation shield for them.

Group cross sections are used as coefficients in the system of 28 differential equations, solution of which defines neutron balance in the core of nuclear reactor. Precision for cross section measurements, requested by applied reactor physics, is determined by the share of delayed neutrons: 0.65% for U^{235}, 0.2% for Pu^{239}, 0.42% for fast breeder reactors. Criticality coefficient, critical mass and breeding ratio of fast neutron reactors must be calculated with precision defined by these values. This requires high statistical precision of TOF measurements.

In spite of good development of the fundamental theory of nuclear interactions, energy dependence curves for neutron cross sections with requested precision can be taken only from the experiment.

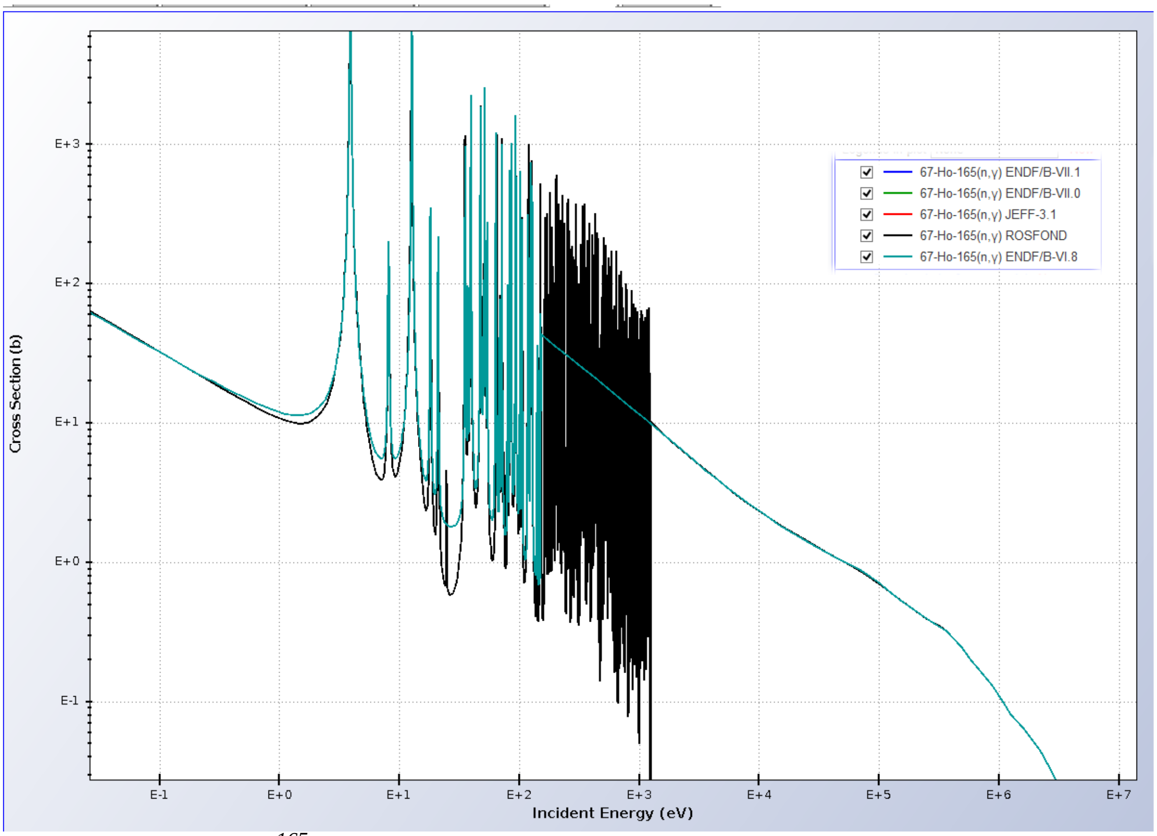

Figure 1. Ho^{165} neutron capture cross section in world data libraries [1].

Ho^{165} capture cross section BNL data [1] are shown of figure 1, in the neutron energy range 0.0253eV<E_n<14 MeV. ENDF/B-VII.1, ENDF/B-VII.0, JEFF-3.1, ROSFOND and ENDF/B-VI.8 data are compared. Upper energy border of resolved resonances in the best world data for Ho165 is 1250 eV. Several disputed resonances were found. Experimental TOF measurements were done to determine, what variant is more true and exact.

2. Experimental equipment

During measurements of Ho^{165} resonance structure, total cross section was measured by 4 Helium-4 based counters SNM-18. Another four counters SNM-18 with 4 atmospheres of He3 were used as monitors of intensity of the pulsed spallation neutron source RADEX. Capture cross section was measured by 40-liter 8-sectional liquid (n,γ) detector of the installation 'INES'. Cadmium beam filter was used in order to cut the recycle neutrons. To determine the background layer of experimental histograms in the low neutron energy area, Mn^{55} beam filter was used.

For determination of the background at high energies, aluminum Al^{27} filter was used. It allowed to determine the background up to neutron energy 140000 eV. Detailed description of the equipment, applied in these measurements, can be found in the sources [2–6].

Accelerator's proton pulse is shown on figure 3. It had duration 250 nanoseconds on half-altitude. Measurements were done during years 2020 and 2021 at the pulsed spallation fast neutron source RADEX, based on tungsten proton beam target of INR RAS 600 MeV proton linac. Metal Ho^{165} radiator pattern was installed inside the 8-sectional liquid (n, γ) detector, at the 50 meter TOF base.

Figure 2. Experimental equipment at 50 meter TOF base, neutrons fly from left to right side.

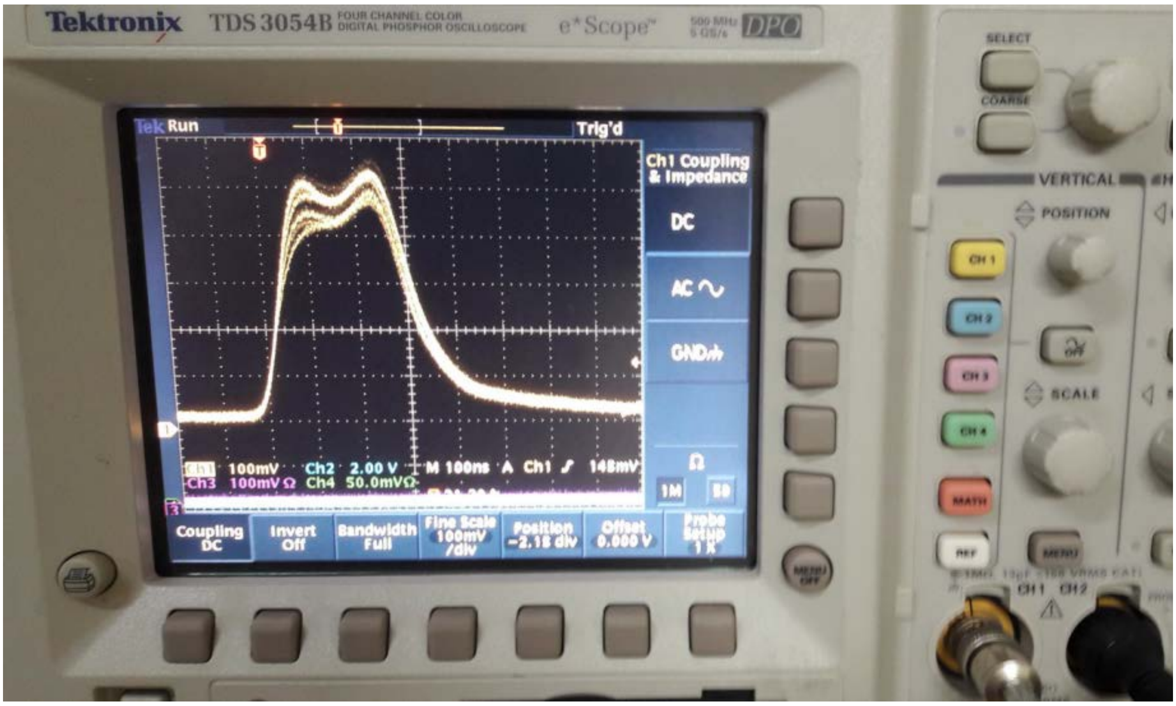

Figure 3. The 250 nanosecond proton beam during turning of the accelerator.

Duration of proton pulse (250 ns), detector's (80 ns) and data accumulation system's (100 ns steps) durations, at 50 meter TOF base provided spectrometer resolution factor: 6 nanoseconds per meter.

3. Analysis of existing world neutron data libraries for Ho^{165}

Comparison of Ho^{165} capture cross section structure in the energy region of resolved resonances showed that ENDF/B-VII.1, ENDF/B-VII.0 and the ROSFOND data for $Ho^{165}(n,\gamma)$ are similar to each other, and have upper border of resolved resonances at neutron energy 1250 eV. Also JEFF-3.1 and ENDF/B-VI.8 are similar to each other and have resolved area up to 152 eV.

Resolution of resonances at higher energies, requires better energy resolution of the spectrometer. At the same time, all differences between data libraries for Ho^{165} are below 152 eV. Between 152 eV and 1250 eV all world data are similar.

As shown on figure 5A, figure 5B and figure 5C, existences of resonances at 24.9 eV; at 75.07 eV; at 120 eV are disputed. Energy position of resonance at 64 eV or at 65.18 eV is disputed. Amplitude of the resonance at 126.9 eV is disputed between four main world data bases of neutron cross sections.

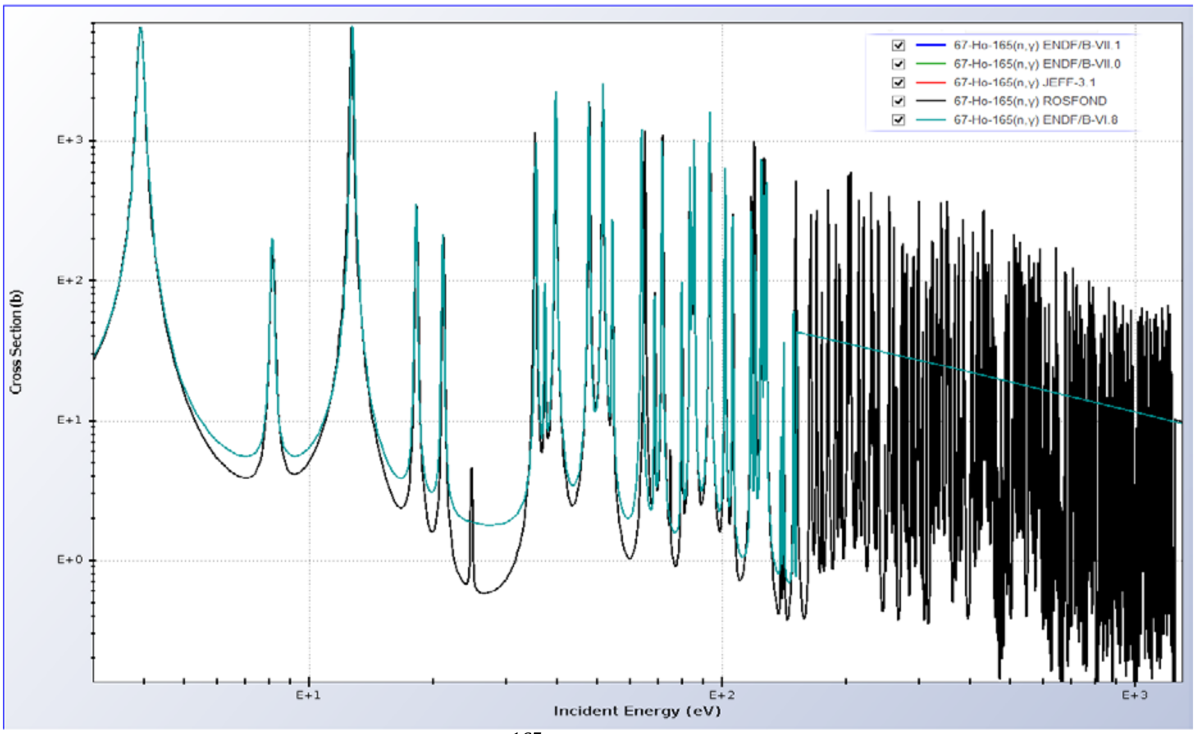

Figure 4. All resolved area of Ho^{165} capture cross section, shown 3 eV<E_n<1250 eV.

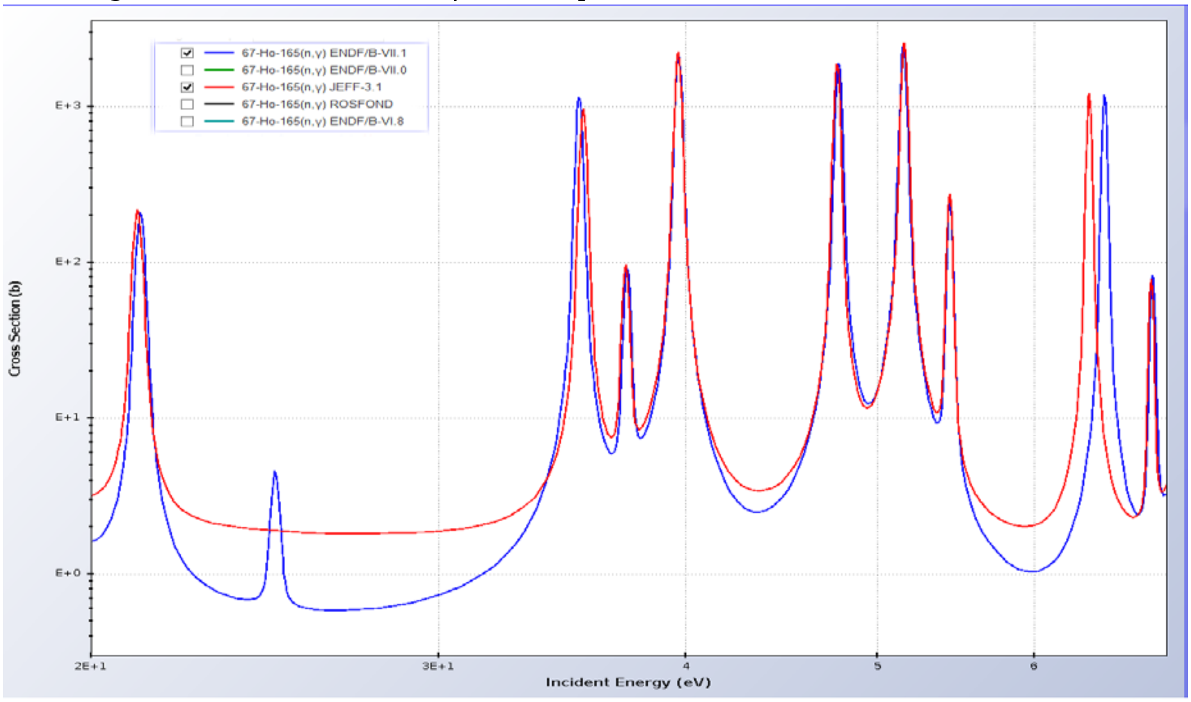

Figure 5A. Ho165 capture cross section debated resonances in the area 20 eV<E_n<70 eV.

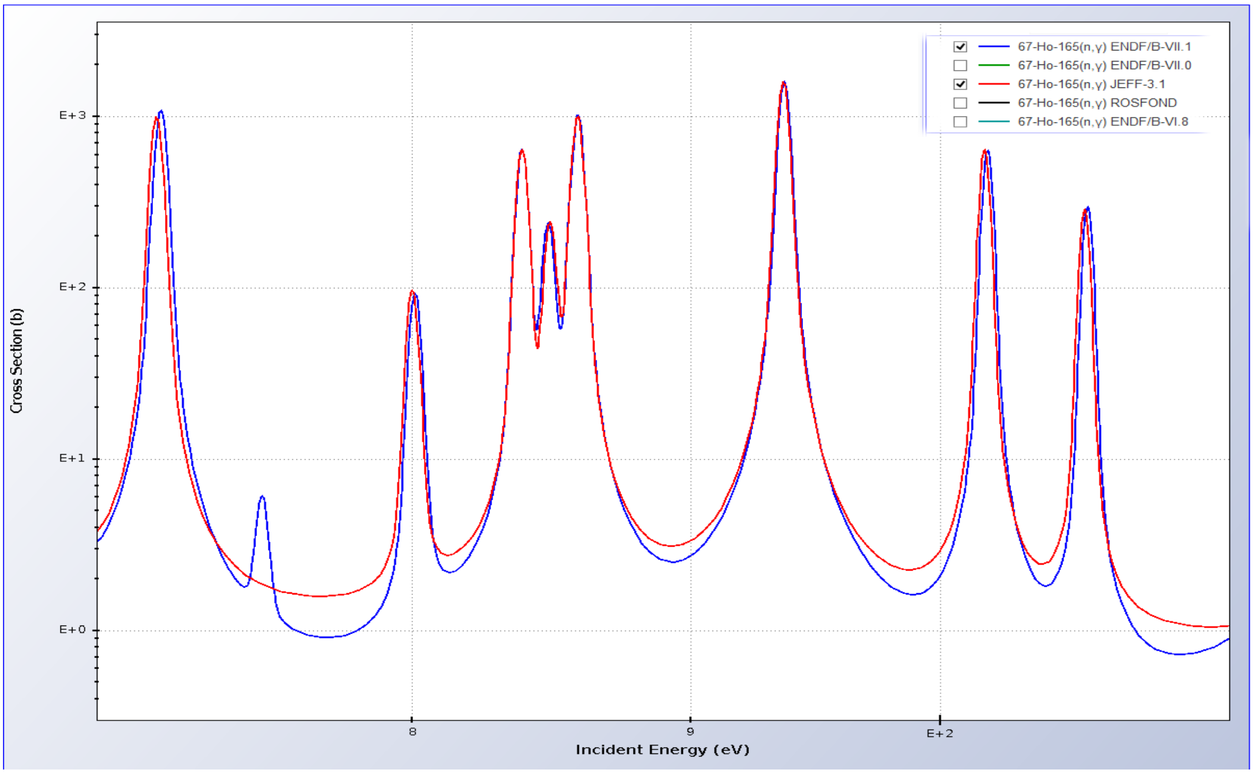

Figure 5B. Ho165 capture cross section debated resonances in the area 70 eV<E_n<120 eV.

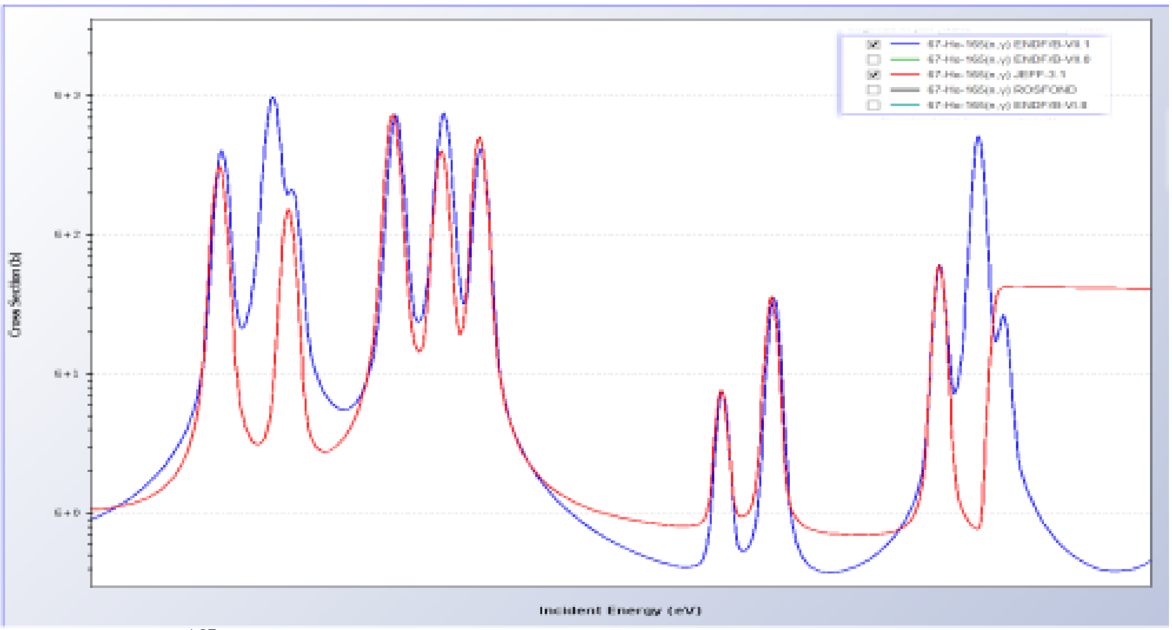

Figure 5C. Ho^{165} capture cross section debated resonances in the area 120 eV<E_n<160 eV.

4. Experimental results for Ho^{165} in the energy area of resolved resonances

Measurements results, collected by data acquisition system, are histogram of 200,000 columns. If we express each channel as one pixel on the computer's screen, wideness of the plot will be equal to 100 screens. We show only some of them.

In figures 6 and 7 we have combined experimental data (upper curve) and BNL ENDF/B-VII.1 data for $Ho^{165}(n,\gamma)$ cross section in barns on right axis, logarithmic scale. We can see that resonances at 24.8 eV, at 65.18 eV and at 75.07 eV exist. Resonance at 64 eV does not exist.

Also we experimentally confirmed, that resonance at 120 eV exists, and that cross section of the 126.9 eV resonance coincidences with the ENDF/B-VII.1 variant. As we can see of figure 8, experimental measurements show good coincidence with the best world data at low and intermediate neutron energies. At high neutron energies, shown on figure 9, resolution 6 nanoseconds per meter of our spectrometer 'INES' based on pulsed neutron source RADEX, allows to distinguish only groups of resonances to get resonance integral value, but some resonances with small energy interval merge.

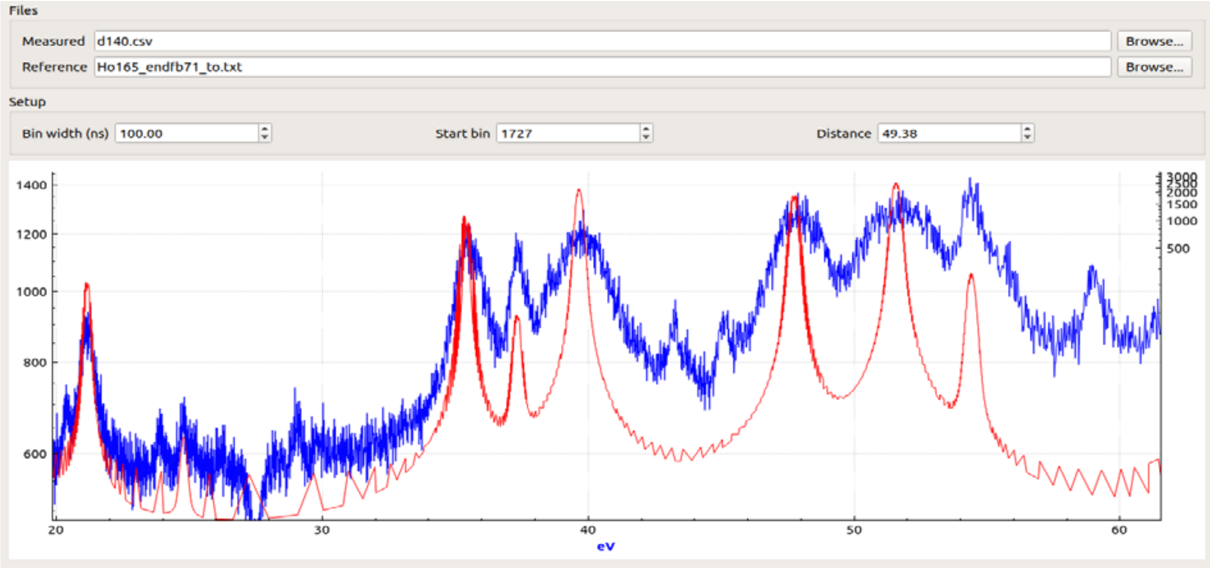

Figure 6. Ho^{165} capture cross section experimental data, 20 eV<E_n<60 eV.

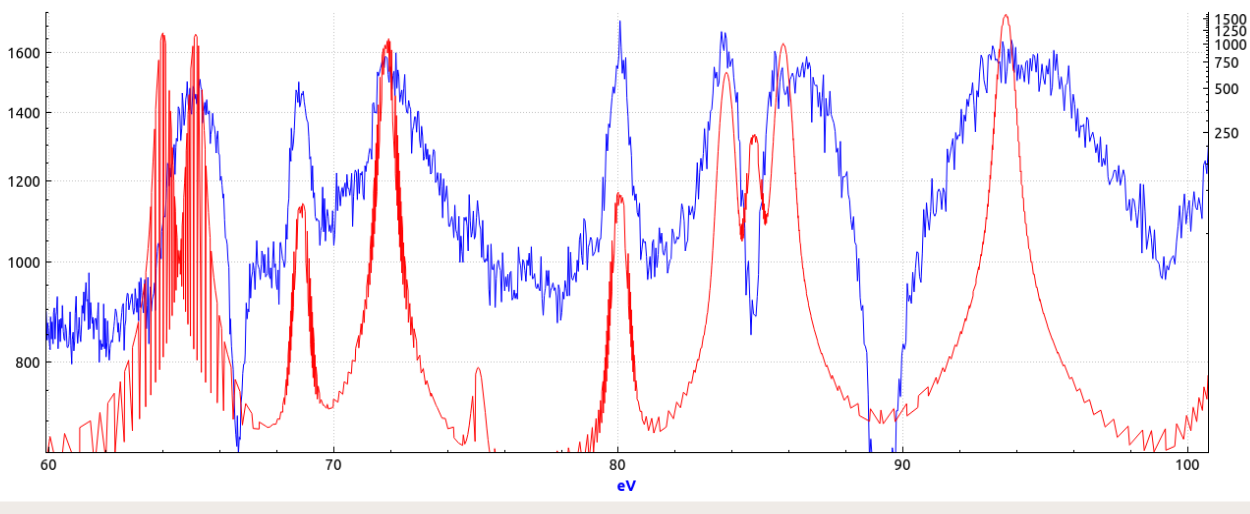

Figure 7. Ho^{165} capture cross section experimental data, 60 eV<E_n<100 eV.

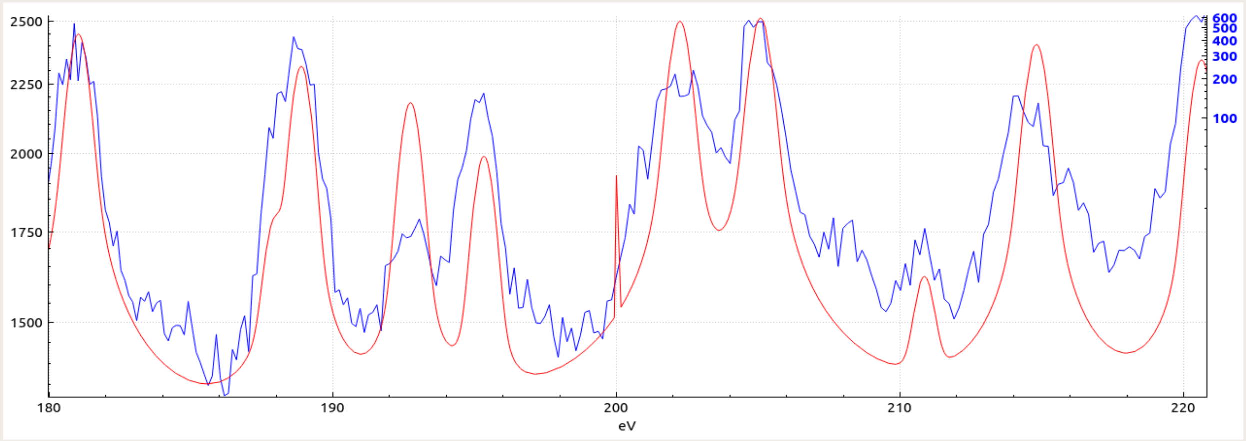

Figure 8. Ho^{165} capture cross section experimental data, 180 eV<E_n<220 eV.

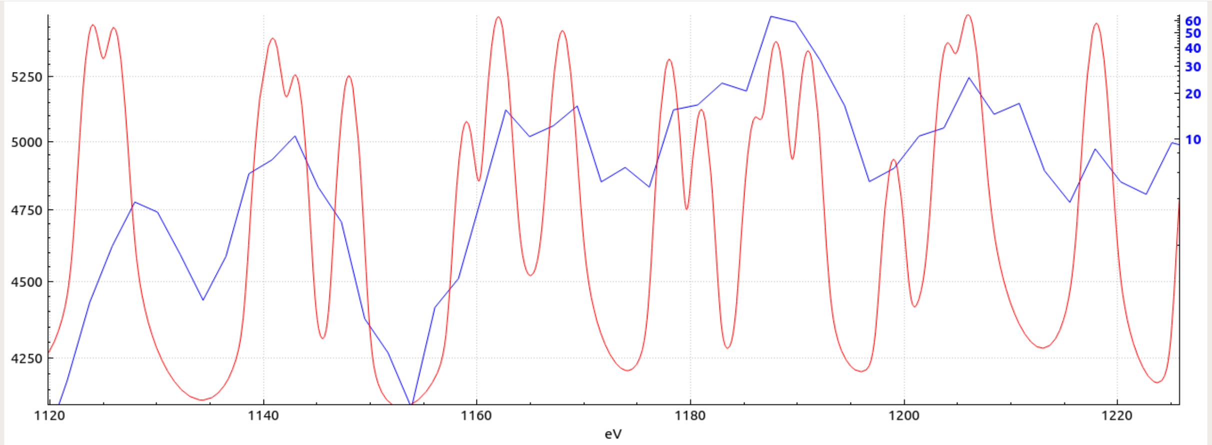

Figure 9. Ho^{165} capture cross section experimental data, 1120 eV<E_n<1200 eV.

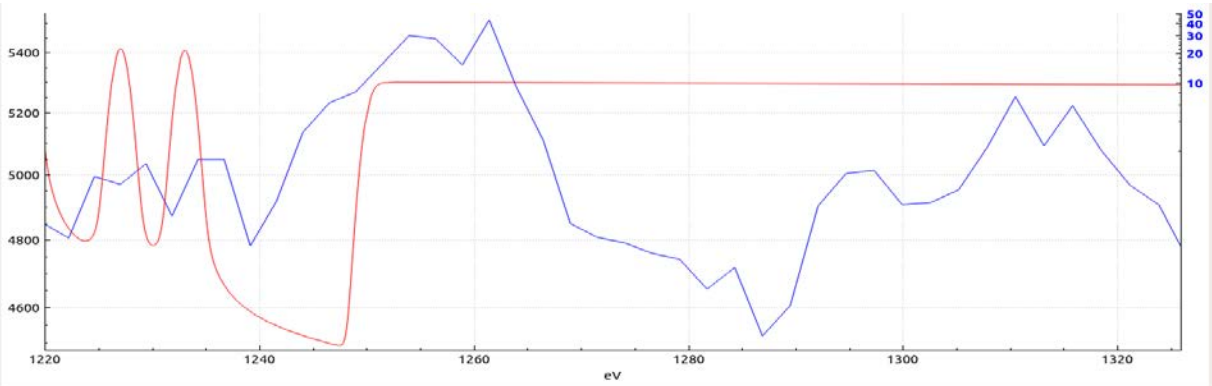

Figure 10. Ho^{165} capture cross section experimental data, 1220 eV<E_n<1320 eV.

In figure 10 we can make sure, that our 'INES' spectrometer's resolution allows to observe Ho^{165} resonance structure, approximately, up to the same 1250 eV, as in the best world data for this isotope. Higher than 1250 eV we observe some histogram peaks, which obviously are groups of resonances.

Above 1250 in the best world data begins interval of unresolved resonances for Ho^{165}, although the Doppler-effect allows to distinguish resonances up to higher energy, what is the subject for future measurements with higher energy resolution.

5. High neutron energy area of unresolved resonances

World TOF spectrometers use pulsed neutron sources of different types: based on electron accelerators, on fission reaction, potentially on inertial thermonuclear synthesis and all their spectrums are well known in world literature [7].

In the case of current measurements on RADEX pulsed neutron source, initial fast particles injected into tungsten target are 247 MeV protons. Spectrum of spallation neutron sources, based on proton accelerators, consists of 2 components. Share of cascade neutrons is 8% of all initial neutrons, they have energy up to energy of protons, i.e. 247 MeV.

Another 92% of neutrons are spallation neutrons, they have average spectrum energy 3 MeV and, compared to fission reaction, less amount neutrons at $E_n < 1$ MeV. Share of delayed neutrons is 10^{-5} and background layer by this factor is very small, if compared to fission reaction as a neutron source where 0.23% for Pu^{239} core and 0.7% for U^{235} core are multiplied to '1-K' factor.

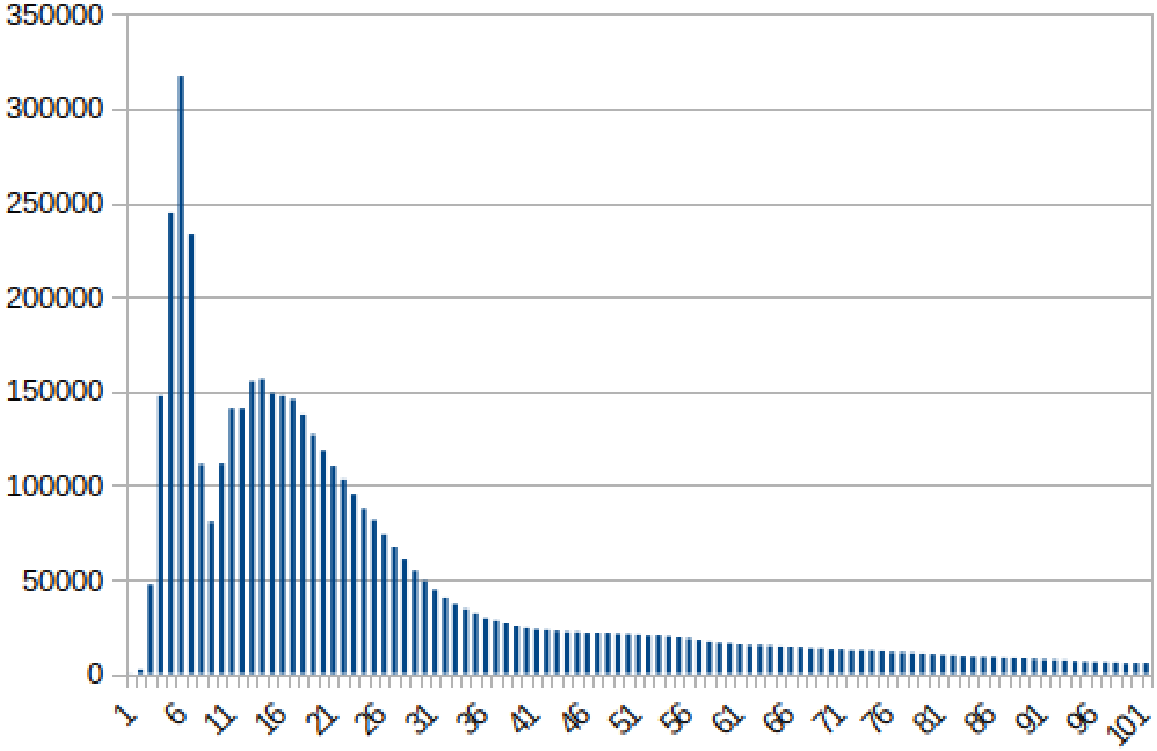

Figure 11. First 100 channels of the histogram.

In the figure 11 first 100 channels of histogram are presented: on horizontal axis channel number with 100 nanoseconds each channel, on vertical axis number of counts per channel. Horizontal axis is especially not transformed into MeV to show, that we distinguish cascade neutrons /first peak/ and spallation neutrons which are second peak, which goes over into Fermi's '1/E' spectrum at lower neutron energies due to availability of H_2O moderator after the tungsten target of RADEX. Also important factor is high value of counts per channel, around 150,000 in the spallation peak. Considering that accuracy of TOF measurements is proportional to $N^{(-0.5)}$, we can achieve precision of cross section measurements up to 0.5% in high energy interval, which is equal to the share of delayed neutrons in fissile materials, and is enough to improve fast breeder reactor multi-group neutron constants.

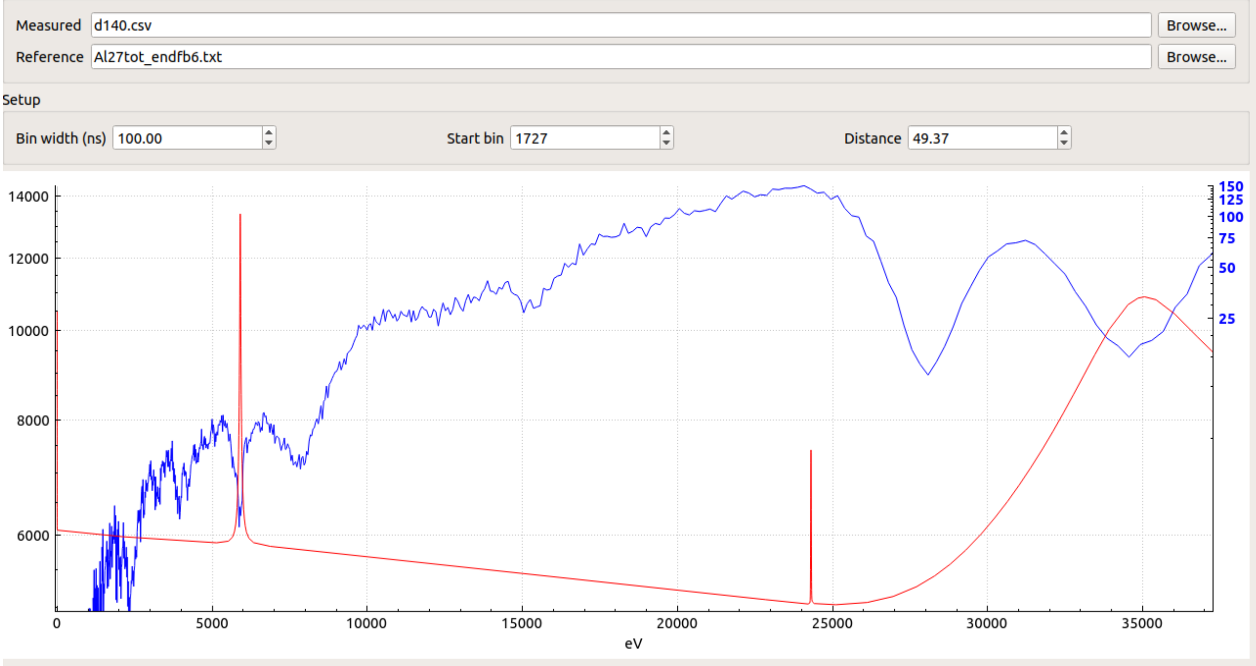

Figure 12. Ho^{165} cross section with Al^{27}filter, 0<E$_n$<37000 eV.

In figure 12, upper curve is Ho^{165} experimental histogram, counts per channel on the left axis. Lower curve is Al^{27} cross section with barns on right axis, Al^{27} resonances at neutron energy 5900 eV and at 35 keV are exactly observable and were used for calibration of the background curve at these energies. Spectrometer energy resolution allows to make measurements on this energy interval, of neutron group cross sections: both for ABBN-78 which has 28 groups. And for group constants based on ABBN-93, like ABBN-RF, which has 299 energy groups and is being developed in Russia for fast breeder reactor calculations.

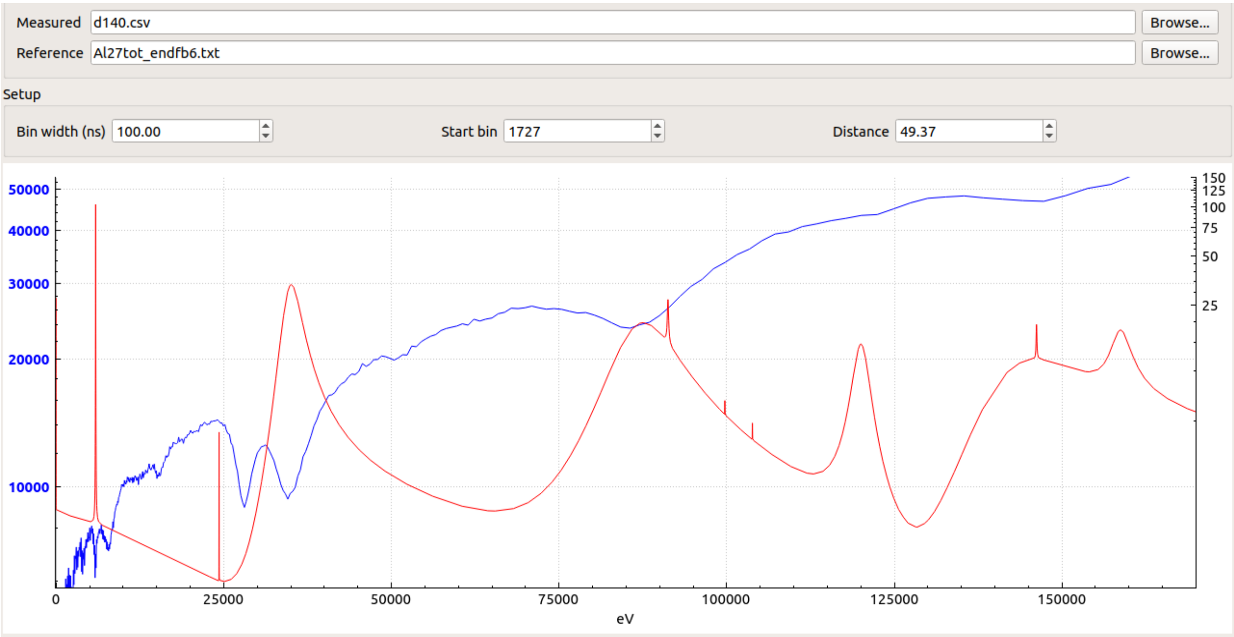

Figure 13. Ho^{165} histogram with Al^{27}filter on lower curve, 0<E$_n$<0.17 MeV.

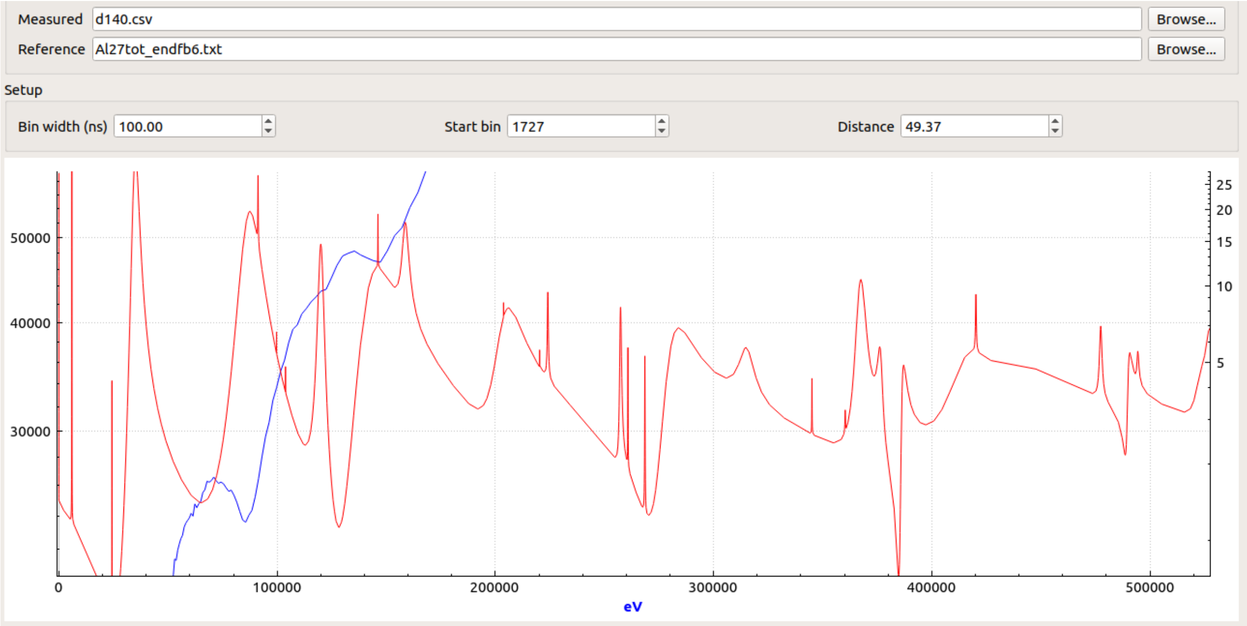

Figure 14. Ho^{165} histogram with Al^{27}filter on lower curve, 0<E_n<0.52 MeV.

Al^{27}(n,total) resonances, used as beam filter, are exactly observable at 5900 eV, at 35000 eV and at 87000 eV on figure 12. Resonance at 147000 eV is also observable on experimental curve on figure 13 and figure 14: counts per channel on left axis for experimental curve, and barns for Al^{27}(n, total) on right axis. This allows to calibrate the background curve in the region 140–170 keV and extrapolate it higher.

Al^{27} resonance at 147000 eV is exactly observable with the scale accepted on figure 14. It's necessary to note, that neutron energy region up to 0.5 MeV includes main part of spectra of fast breeder reactors. Initial fission spectrum has average energy 2 MeV and most probable energy 650 keV. In fast spectrum reactor, due to inelastic and elastic scattering on construction isotopes of the reactor's core, average spectrum energy is shifted to 150 keV. Resonance at 0.14 MeV of Al^{27} proves, that we can measure such energies on the INES TOF spectrometer of RADEX pulsed neutron source. Also, new method for background determination in the MeV region is currently investigated.

6. Conclusion

Ho^{165} capture resonances were experimentally confirmed at 24.8 eV; at 65.18 eV; at 75.07 eV; at 120 eV. Amplitude value for 126.9 eV resonance coincidences with ENDF/B-VII.1 data file.

Energy resolution of the spectrometer INES, based on neutron beam of the INR RAS pulsed neutron source RADEX, allows to observe all resolved area of Ho^{165} capture cross section. Up to 1250 eV, according to the best world data files.

At resolution 6 nanoseconds per meter we achieved separation between cascade neutrons and spallation neutrons. In the 'figure 11' first 100 histogram channels of (n,γ) detector are shown, 100 ns each step, energy of minimum point between components is 14 MeV.

Energy resolution of modern TOF spectrometers allows to make measurements in all practically important neutron energy region: 0.0253 eV < E_n < 14 MeV.

It allows to resolve the resonance structure, of all 286 stable nuclides, up to energy, where Doppler-effect makes intersection of neighbor resonances at the temperatures of the core of nuclear reactors and radiation shield.

The same high energy resolution is necessary to make possible measurements in wide energy groups for multi-group neutron reactor constants in the practically important energy area of hundreds keV and several MeV.

7. Literature and cited sources

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