

UTILIZING THE SLOWING-DOWN-TIME TECHNIQUE FOR BENCHMARKING NEUTRON THERMALIZATION IN GRAPHITE

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Graphite is the moderator/reflector in the Very High Temperature Reactor (VHTR) concept of Generation IV reactors. As a thermal reactor, the prediction of the thermal neutron spectrum in the VHTR is dependent on the accuracy of the thermal neutron scattering libraries of graphite. In recent years, work has been on-going to benchmark and validate neutron thermalization in “reactor grade” graphite. Monte Carlo simulations using the MCNP5 code were used to design a pulsed neutron slowing-down-time experiment and to investigate neutron slowing down and thermalization in graphite at temperatures relevant to VHTR operation. The unique aspect of this experiment is its ability to observe the behavior of neutrons throughout an energy range extending from the source energy to energies in the thermal range. In its current form, the experiment is designed and implemented at the Oak Ridge Electron Linear Accelerator (ORELA). ORELA neutron pulses (130 Hz) are injected into a 70cm x 70cm x 70cm graphite assembly. A furnace system that surrounds the assembly and is capable of heating the graphite to a centerline temperature of 1200 K has been designed and built. A system based on U-235 fission chambers and Li-6 scintillation detectors is used in the experiment. This system is coupled to multi-channel scaling instrumentation and is designed for the detection of leakage neutrons as a function of the slowing-down-time (i.e., time after the pulse). To ensure the accuracy of the experiment, careful assessment was performed of the impact of background noise (due to room return neutrons) and pulse-to-pulse overlap on the measurement. Therefore, the entire setup is surrounded by borated polyethylene shields. As the basis for the benchmark, the calculated time dependent reaction rates in the detectors (using the MCNP code and its associated ENDF-B/VI thermal neutron scattering libraries) are compared to measured values.

Introduction

Since the early days of nuclear power, graphite has played an important role as a neutron moderator. In “thermal” reactors, high energy neutrons that are produced in the fission reaction lose their energy through elastic scattering with the carbon atoms, which increases the probability of their absorption in the fuel to cause further fission and allows for the fission chain process to occur. As neutrons enter the thermal region (neutron energy ≤ 1 eV), the effect of the bonding of the moderator atoms (or molecules) becomes important in defining the thermal neutron energy spectrum. In these reactors, accurate knowledge of the thermal neutron energy spectrum affects the accuracy of the core design calculations, which influences the safety and economic (i.e., fuel utilization) aspects of the reactor. Consequently, accurate determination of the thermal neutron scattering cross sections of the moderator becomes a necessity.

To date, the thermal neutron scattering libraries for graphite have been based on models and theoretical techniques that were developed in the fifties and sixties of the twentieth century [1]. The basic approach that was followed is to fit a proposed dynamic model of the graphite lattice to experimental results. The outcome of this exercise gives the vibrational information (i.e., phonon spectrum) for graphite. The phonon spectrum can then be used in computer codes such as NJOY (and its LEAPR module) to generate the thermal neutron scattering cross sections [2]. However, nowadays advances in computational power make it possible to implement more

fundamental theoretical and computational approaches to achieve this objective [3,4,5].

To benchmark cross section libraries, the Slowing-Down-Time (SDT) method utilizes the time and energy behavior of bursts of neutrons injected into a moderating assembly [6,7,8]. The fast neutrons injected into the graphite pile will begin to slow down by inelastic (if energetically allowed) and elastic scattering until they reach energies comparable to the thermal energies of the moderator atoms. The main advantage of the Slowing-Down-Time technique is that the evolution (in time) of the neutron energy spectrum in the moderator from the source energy down to the thermal energy range can be monitored. The fast neutron region in a measured time spectrum can be used to normalize computational predictions. Because the fast neutron cross section of graphite is considered well-known, accurate computational predictions in the fast neutron region in the time spectrum are considered tractable. The comparison between measurements and normalized calculations can be made not only to assess the shape of the thermal neutron spectrum but also the amplitude. Additionally, the basic data collected during this experiment can be directly compared to computational predictions without the need for extensive processing that can introduce undesirable uncertainties. Those features are considered an advantage over the more traditional Time-Of-Flight method.

Design of the Graphite SDT Experiment

The Monte Carlo code MCNP version 5 and its ENDF/B-VI cross-section libraries [9] were used in this experiment to perform neutron transport calculations in order to study the slowing down of neutrons in a graphite moderator. The basic design of the experiment is given elsewhere [7,8]. The Oak Ridge Electron Linear Accelerator (ORELA) facility is the neutron source for this experiment [10]. The source neutron energy spectrum, despite its spread, is a fast neutron spectrum peaking around 600 keV. The experiment (Figures 1 and 2), which was setup in the ORELA basement, involved a graphite assembly, a heating system (furnace and its controller), a shielding system, a temperature monitoring system, a gas regulation system and a data acquisition system.

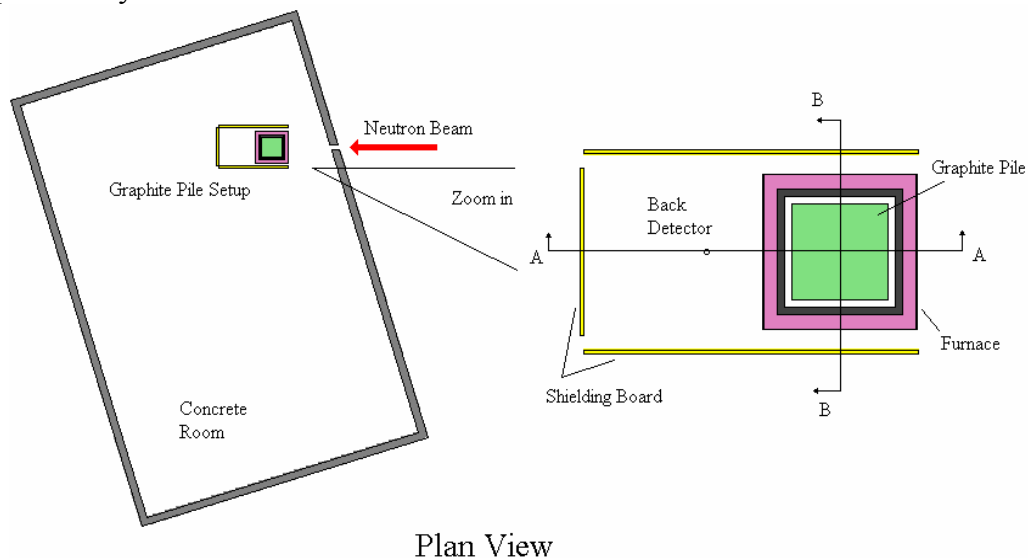


Fig. 1. Top view of the experimental setup.

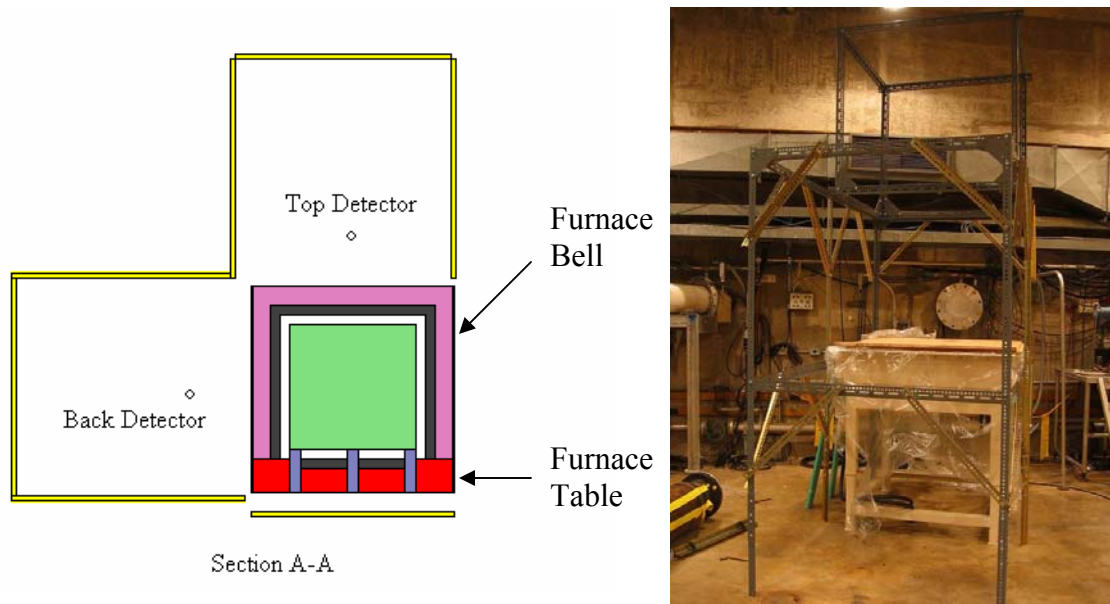


Fig. 2. A schematic and picture of the experimental setup.

To study thermal neutron scattering in graphite at temperatures extending up to 1200K, which is a temperature range relevant to Very High Temperature Reactors (VHTR), a furnace is introduced that provides heating ability up to such temperatures. The furnace consists of two detachable parts; a furnace bell and an associated base. For room temperature measurements, the furnace bell can be removed from the setup. The graphite assembly is placed on the furnace table. The size of the assembly can be 50x50x50 cm, 70x70x70 cm or 100x100x100 cm with the furnace bell opened (room temperature), and 70x70x70 cm with the bell closed (high temperature). There are 10 thermocouples distributed inside the graphite assembly to monitor the temperature in real time. As a safety precaution, the gas regulation system introduces nitrogen cover gas into the furnace to purge oxygen and keep an over pressure in the furnace. To reduce room-return neutrons, polyethylene shielding boards that are loaded with 5 weight % boron surround the entire setup. The left panel in Fig. 2 illustrates the furnace table, the frame structure that holds the shielding boards and detectors, and the neutron beam port. The data acquisition system is shown in Fig. 3. The detector could be located either on top or the back of the graphite assembly. The Slowing-Down-Time spectra are recorded using a multi-channel-scaler (MCS) system. In addition, the neutron pulse height spectra are monitored using a multi-channel analyzer (MCA). All the data acquisition and monitoring units in the ORELA basement are remotely controlled using computers in the data acquisition room on the 2nd floor through an Ethernet connection.

Since the ORELA neutron source is pulsed, every neutron pulse will produce a signal in the detector, which will be accumulated in the MCS. The signal produced by each pulse extends for 0.03 seconds. If the pulse repetition rate is high, a neutron pulse can arrive before the signal corresponding to the previous pulse completely dies away. Consequently, pulse overlap effects can occur. Figure 4 shows the expected MCS time spectrum, using a Pu-239 fission detector, at the typical ORELA source frequencies of 525 and 130 pulses/second. At a frequency of 525 pulses/second a very large pulse overlap effect is observed. While at a frequency of 130 pulses/second the pulse overlap effect is negligible. Both cases are compared to the time

spectrum without overlap effect. Clearly, the lower frequency is preferred. The percentage effect of pulse overlap at the frequency of 130 pulses/second is less than 2% in the time period 10^{-4} to 3×10^{-3} second, which is relevant for observing thermal neutrons.

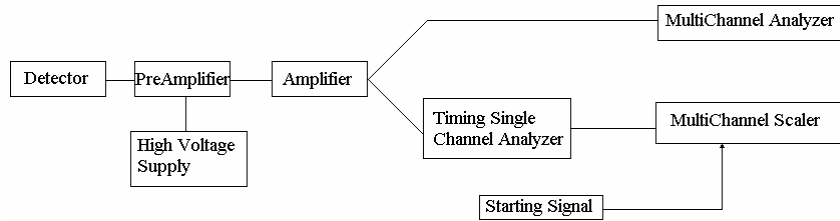


Fig. 3. A schematic of the data acquisition electronics.

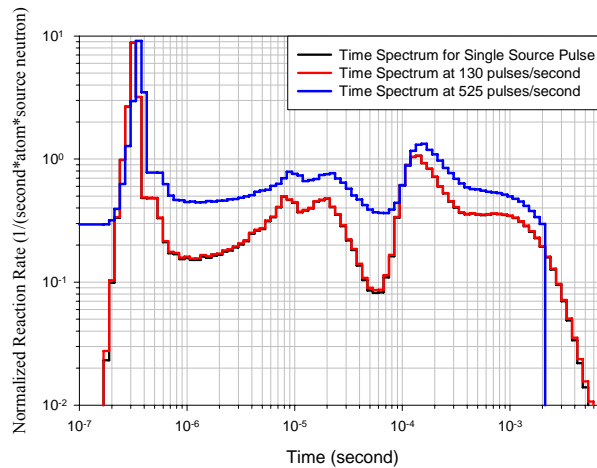


Fig. 4. Pulse overlap effects at a frequency of 130 and 525 pulses/second on the Pu-239 time spectrum.

Sensitivity of the SDT Experiment

In this work, the MCNP5 code and its point detector (F5) tally feature were used in the design of the SDT experiment and the prediction of the collected time spectra. As part of the design process, MCNP5 was modified to allow applying the differential operator perturbation method to the point-detector tally [9]. The modified code was used to analyze the sensitivity of the SDT spectrum to the graphite thermal neutron scattering cross sections. Figure 5 shows the impact on the predicted time spectrum of a 3% increase in the total thermal neutron cross-section. In addition, the existence of the furnace and the concrete room is shown to reduce the sensitivity, while introducing the shielding system results in recovering some of the sensitivity lost due to room return effects. Figure 5 also shows that assuming that the real graphite thermal neutron total scattering cross section is 3% higher than in the data libraries, the measured time spectrum will be 8% lower than the calculated one in the thermal neutron region around 0.5 milliseconds.

In reality, the current thermal neutron scattering cross section for graphite in the ENDF/ B-VII library [11] shows a notable discrepancy in comparison to the measured data below the Bragg-cutoff energy, as shown in Fig. 6. This discrepancy is remedied using techniques developed at North Carolina State University (NCSU). In particular, ab initio methods were combined with lattice dynamics calculations to generate the data needed for calculating the thermal scattering cross sections for graphite, while including the coherent inelastic scattering

component that is usually neglected in the NJOY formulation [3,4,5]. This data library is designated as NCSU-1P. As it can be seen in Fig. 6, both the ENDF/B-VII and the NCSU-1P libraries agree with measured data above the Bragg-cutoff energy. Below the Bragg-cutoff energy, the NCSU-1P library matches the results of measurements in pyrolytic graphite [8,12].

Neglecting the neutron absorption cross section of graphite, the total thermal scattering cross section is equal to the sum of the elastic and inelastic scattering cross sections. A thermal neutron can be up scattered (gain energy) or down scattered (lose energy). Below the Bragg-cutoff energy, elastic scattering is impossible and the total cross section is equal to the inelastic cross section. The inelastic scattering cross section data in the ENDF/B-VII library are obviously lower than the measurements. Consequently, above the Bragg-cutoff energy, the agreement between the total cross section in ENDF/B-VII and the measurements results from underestimated inelastic and overestimated elastic cross sections. This can be verified by the comparison between the ENDF/B-VII and NCSU-1P libraries elastic and inelastic cross sections separately as seen in the right panel of Fig. 6. It should be noted that the graphite thermal neutron scattering cross sections in the ENDF/B-VI and ENDF/B-VII libraries are identical.

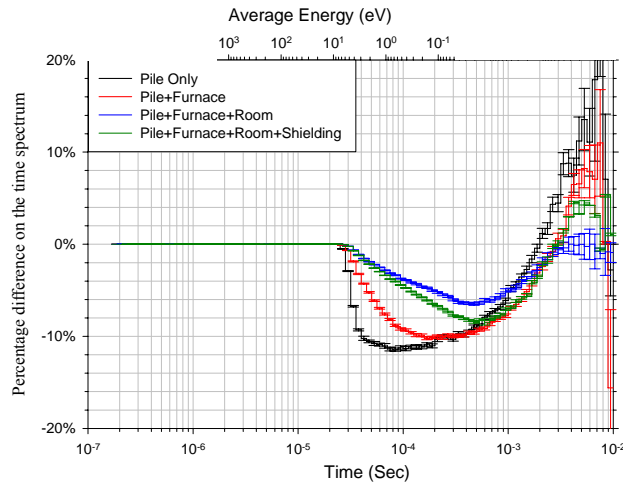


Fig. 5. The perturbation calculation of the time spectra due to a +3% increase in the graphite total thermal neutron cross section for the back detector at room temperature.

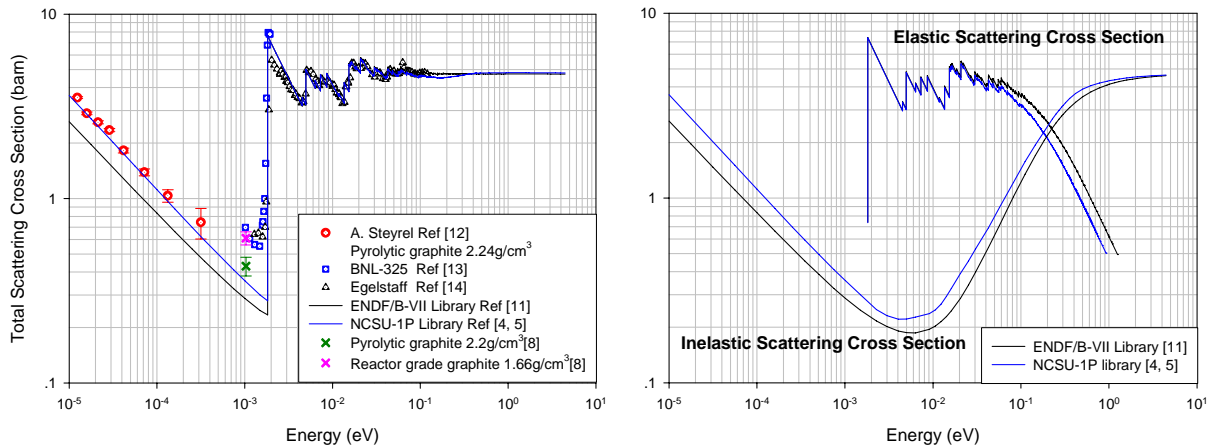


Fig. 6. Graphite total, elastic and inelastic scattering cross section libraries.

The modified MCNP5 code was used to calculate the impact on the time spectrum of a

perturbation of 5% of the total value of the elastic and inelastic cross sections separately in the energy region of 0.01eV to 1eV, which is the relevant energy region for the SDT experiment. The results are shown in Fig. 7. As it can be seen, the same perturbations of the elastic and inelastic cross section separately will give different results. Therefore, the combined result, shown as the black solid line in Fig. 7, gives an indication of the difference in the time spectrum due to the combination of underestimated inelastic and overestimated elastic thermal neutron scattering cross sections. The difference between the perturbation calculations of 5% increase in inelastic and elastic cross sections can be explained by the fact that inelastic scattering allows neutron energy transfer while elastic scattering does not.

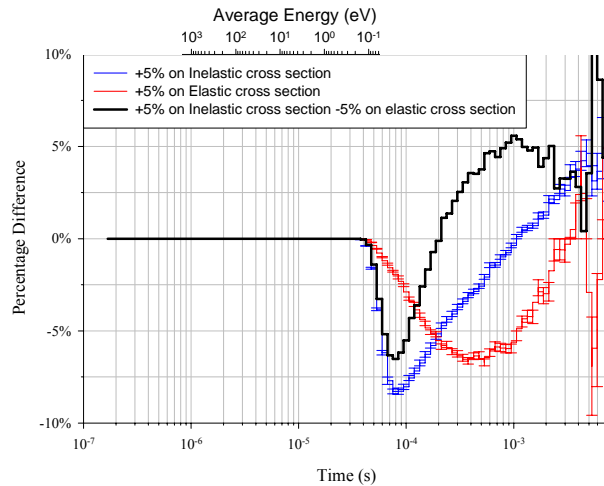


Fig. 7. Back detector perturbation calculations based on a +5% increase in the inelastic and elastic scattering cross sections. The solid black line is the combination of a 5% increase of inelastic and 5% decrease of elastic cross sections.

Using the NCSU-1P library, a time spectrum was calculated using the MCNP5 code and compared to the spectrum calculated based on ENDF/B-VII library. Figure 8 shows that the difference between the two spectra is as large as 5%. Notice that in this case the libraries discussed above are for crystalline graphite. However, in VHTR applications and in the ORELA measurements, reactor grade graphite will be utilized which has a different structure than crystalline graphite.

Slowing-Down-Time measurements

The ORELA SDT measurements are performed using a Li-6 glass scintillator detector placed at the back and top of the graphite 70x70x70 cm assembly. The thickness of the lithium glass is 1mm and the diameter is 3 inches. The density is approximately 2.5g/cm³. The total lithium in the detector is 6.6% and the Li-6 enrichment is 95%. The detailed settings of the data acquisition system are shown in Table 1.

Figure 9 illustrates the measured data (top detector) in comparison to the spectra calculated by the MCNP5 code based on the ENDF/B-VI library. The calculated spectra were normalized in order to have the same total counts in the fast neutron region, 9.46x10⁻⁶ to 4.22x10⁻⁵ second, as in the measured spectra. This is considered to be reasonable because the fast neutron cross sections of graphite in the ENDF/B-VI library are free carbon atom cross sections which are well measured and tested. Therefore, it is expected that the time spectra in the fast neutron region

should be accurately predicted by the MCNP calculations. As it can be seen in Figure 9, the calculations agree with the measured spectra in the time region earlier than approximately 7×10^{-5} seconds, i.e., the neutron energy is above 4eV. However, the calculated spectrum does not agree with the measured spectrum in the time region earlier than 7×10^{-6} seconds. To avoid the gamma-ray flash that accompanies the ORELA neutron burst, the detector was placed at the top location perpendicular to the path of the neutron beam. However, in general, the gamma-ray background was eliminated using a discriminating window in the timing Single Channel Analyzer, which is possible due to the large Q-value (4.78MeV) of the (n, α) reaction of Li-6.

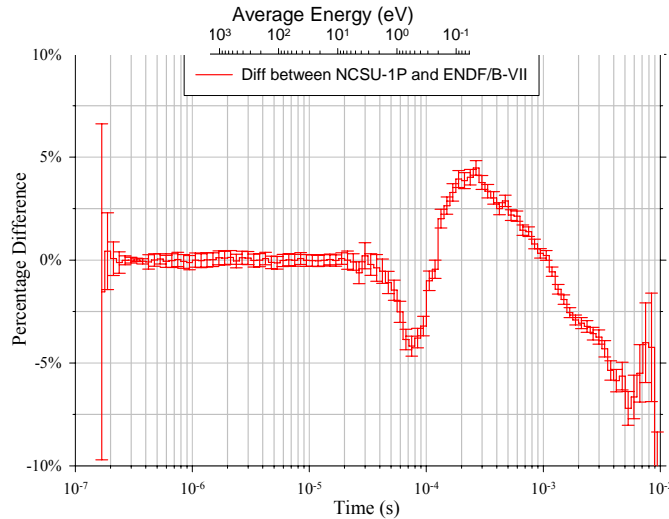


Figure 8. The difference between the time spectra of the back detector based on the NCSU-1P and ENDF/B-VII libraries.

Table 1 Parameters in the data acquisition system

Detector	Li-6 glass
Measurement time	1 hour
High Voltage	850V
Amplifier gain	0.55×100
Amplifier shaping time	$1 \mu\text{s}$
Timing SCA window	0.9V ~ 4V
Timing SCA delay time	$6 \mu\text{s}$
MCS channels	8192
MCS dwell time	$0.9 \mu\text{s}$
MCA channels	2048

Neutrons slow down into the thermal energy range around 7×10^{-5} seconds after the injection of the source pulse into the graphite assembly. In the thermal region, the calculated time spectra begin to diverge from the measurements. As time progresses the difference between the measured and calculated time spectra is large and is possibly attributed to ORELA thermal neutrons that were underestimated in the simulation of the source. To assess this effect, future measurements are planned using Cd and/or boron loaded filters to remove thermal and

epithermal neutrons from the pulse. This would enable isolating the impact of low energy neutrons on the time spectrum.

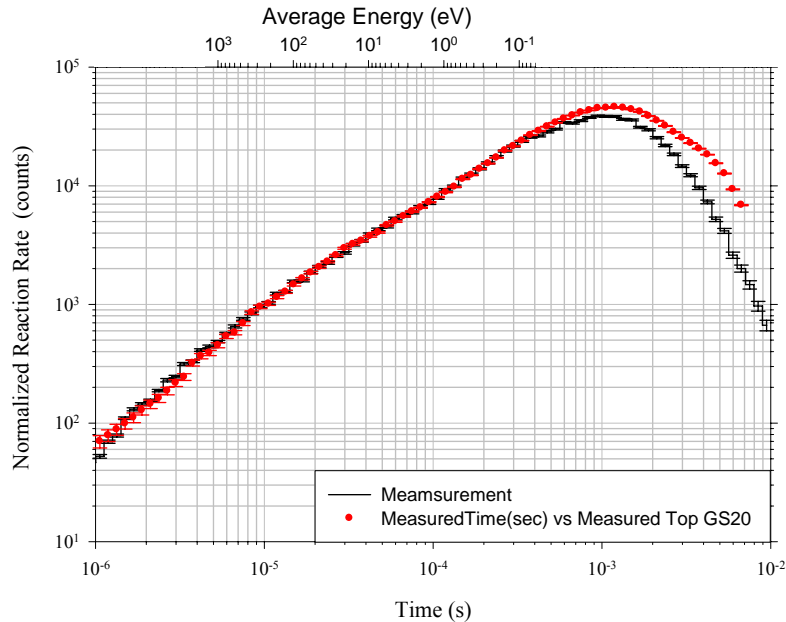


Fig. 9. Simulated and measured time spectra of the Li-6 glass scintillator on the top of the graphite assembly.

Conclusions

A slowing-down-time experiment in graphite has been designed and is in its first stages of execution at the ORELA facility in Oak Ridge National Laboratory. The experiment aims at benchmarking the slowing down and thermalization of neutrons in graphite at temperatures extending from room temperature to approximately 1200K. This is considered to be a relevant temperature range for the VHTR. Measurements performed so far have shown significant deviations from computational predications in the thermal energy range. However, better assessment is needed of the thermal neutron contamination in the ORELA source to improve the understanding of the background of the measurement. Consequently, future measurements are planned using Cd and/or boron loaded filters to remove thermal and epithermal neutrons from the source pulse.

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