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MONTE CARLO SIMULATIONS FOR TIME-OF-FLIGHT EPITHERMAL NEUTRON ACTIVATION ANALYSIS FOR ISOTOPIC SIGNATURES

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ABSTRACT
Epithermal neutron activation analysis (ENAA) is a technique that can leverage unique isotopic resonances of interest. An epithermal neutron time-of-flight (TOF) facility is being developed at the Penn State Breazeale Reactor (PSBR) for use as a nuclear material verification and elemental/isotopic characterization tool. This new facility will implement a novel mechanical chopper to pulse epithermal neutrons up to 40 eV with an energy resolution of approximately 2%. Previous work has focused on the design of the mechanical chopper that will create the desired source of nearly monochromatic epithermal neutrons. This work focuses on analytical calculations to quantify the neutron chopper's performance, as well as on Monte Carlo simulations that have been used to evaluate the chopper design and its utility for TOF and ENAA measurements. Specifically, dynamic simulations have been modeled by discretizing independent MCNP6.2 simulations in time to model the expected performance of the mechanical neutron chopper.

INTRODUCTION
There is a need for a fast, non-destructive technique to monitor nuclear material throughout the fuel cycle. Elemental and isotopic information can be extracted through thermal and epithermal neutron interrogation. Unique signatures can be utilized to enhance the ability to monitor and verify nuclear-material activities. ENAA is a technique that can leverage unique isotopic resonances of interest. Epithermal neutron activation can allow for more information to be extracted compared to thermal or full reactor spectrum activation analysis [1]. To maximize the ENAA performance, it is desirable to have a monochromatic source of neutrons for activation analysis to reduce activation of undesirable nuclei [2]. Current mechanical neutron choppers and velocity selectors are unable to monochromatize neutron pulses above a few eV. This is due to their inability to move sufficient absorbing mass at the velocities required for a desired energy resolution. An epithermal neutron TOF facility is being designed for the TRIGA Mark-III research reactor at Penn State. This facility will implement a mechanical neutron chopper to pulse epithermal neutrons up to 40 eV with energy resolution of approximately 1 eV. The energy resolution improves for neutrons of lower energies.

BACKGROUND
Many isotopes exhibit unique absorption resonances in the epithermal neutron energy range. Thermal and cold neutrons are more commonly used to extract elemental information from large contrasts of scattering or absorption cross-sections where quantitative measurements can provide bulk sample information. Epithermal neutrons can provide additional sample information due to unique, narrow resonances in isotopic cross-sections.

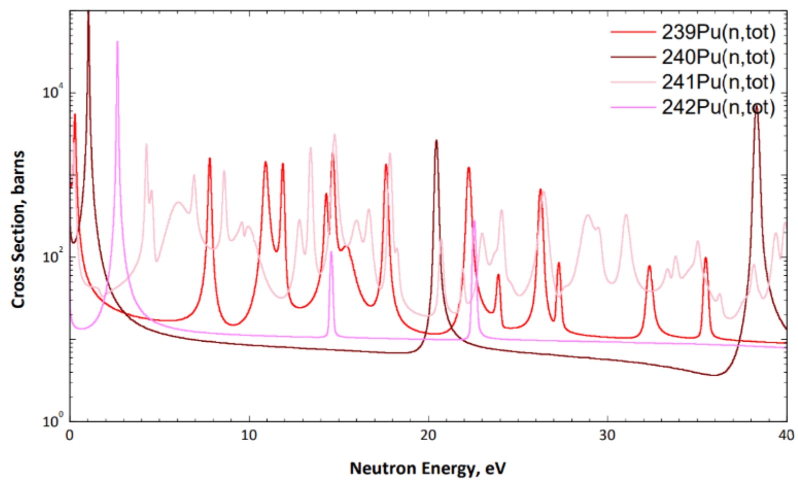

Figure 1. ENDF/B-VII.1 total neutron cross-sections for isotopes of plutonium between 0.1 and 40 eV [3].

Figure 1 shows that many non-overlapping resonances exist in the desired neutron energy range. The cross-sections at resonant energies can also span orders of magnitude across isotopes. Interrogation of a sample with neutrons in the epithermal energy range of interest can be utilized as a non-destructive analysis tool to map elemental and isotopic composition of a given sample provided it has resonances in this energy range. For this reason, neutron resonance transmission analysis (NRTA) also has applications in nuclear safeguards and verification of treaties [4]. Interrogation with epithermal neutrons can also result in the emission of gamma rays unique to the target isotope. The prompt gamma radiation measured during activation can be used to quantify trace elements or isotopes present in the sample with high sensitivity. This technique is known as the neutron resonance capture analysis (NRCA). There are a few disadvantages to this method compared to NRTA. One disadvantage is that the measurement of prompt gamma rays will require a longer interrogation time to obtain the same uncertainties on the measurements. The signal for NRTA is the incident neutrons arriving from a pulsed neutron source. However, the signal for NRCA is the gammas detected from activation. Therefore, there will inherently be a lower intensity of gammas produced compared to the original neutrons, and the gammas are not collimated. The second disadvantage is that the NRCA method requires the neutron beam to scan the sample to achieve spatial resolution. The spatial resolution is directly correlated to the size of the incident neutron beam [5]. The NRTA method can spatially resolve material composition based on the active area of the pixelated detector. The neutron transmission observed in the pixelated detector can be compared with and without a sample for normalization thus eliminating the need to scan the sample. The TOF facility being designed at the PSBR will combine TOF measurements with prompt gamma analysis to elementally/isotopically map a sample, while quantifying trace atoms present for a verification of nuclear material or nuclear material-related activities. A pixelated neutron detector for the TOF measurements will allow for elemental and isotopic mapping, while the prompt gamma analysis can quantify trace impurities.

ANALYTICAL CALCULATIONS

The design of the mechanical neutron chopper has been optimized to maximize the pulse intensity and the energy resolution for a given set of system parameters. Deterministic calculations were used to create Monte Carlo simulations for a stochastic validation of the neutronics performance.

Energy Resolution

The energy resolution of a neutron pulse can be related to the uncertainty in its kinetic energy by:

$$\frac{\delta E}{E} = \sqrt{\left(2\frac{\delta L}{L}\right)^2 + \left(2\frac{\delta t}{t}\right)^2} \tag{1}$$

Assuming the uncertainty in length is negligible for a given flight path [6]:

$$\frac{\delta E}{E} = 2\frac{\delta t}{t} \tag{2}$$

Due to a finite shutter time of the mechanical chopper, and a fixed flight path, the energy resolution can be defined in terms of the pulse duration and TOF of a nominal neutron energy as:

$$\frac{\delta E}{E} = 2\left(\frac{Shutter\ time}{TOF}\right) \tag{3}$$

Neutron Intensity

The theoretical neutron intensity has been determined analytically as a function of chopper and source parameters. For a given source and mechanical chopper system with any set of geometrical parameters and cyclic frequency, the resulting neutron intensity can be defined as the following:

$$Intensity\left[\frac{n}{kW*s}\right] = source\ \left[\frac{n}{cm^2*s*kW}\right]*shutter\ time[s]\ *\omega\left[\frac{1}{s}\right]*slit\ area[cm^2] \tag{4}$$

The shutter time is directly related to the slit width and velocity of the mechanical chopper. This parameter dictates the energy resolution for a given nominal neutron energy over a fixed flight path. Geometrical parameters dictate the slit area, and it is assumed that this area of exposure is constant for the full duration of the pulse. Mechanical limitations for a given set of parameters result in a maximum achievable velocity of the chopper system. These system parameters have been optimized to maximize the resulting neutron intensity.

MONTE CARLO SIMULATIONS

A Monte Carlo simulation tool has been developed to evaluate the neutronics performance of the proposed mechanical neutron chopper. This tool is used to optimize all relevant system parameters of the mechanical chopper. Utilizing the optimized geometry and velocity of the chopper that has been analytically determined, the tool updates the input files for MCNP6.2 accordingly. In the next step, it performs discretized chopper rotations and runs neutronics simulations to mimic the dynamic system behavior. In addition, the Monte Carlo simulations were utilized to stochastically validate the analytical calculations such as the energy resolution. These simulations have been extended from the previously defined piston-type geometry to evaluate the performance of a series of disc choppers that will be aligned on a single shaft. Two systems of disc choppers will be utilized to create a single velocity selector: One at the start and one at the end of the flight path.

Energy Resolution and Intensity

Monte Carlo simulations have been used to evaluate the performance of two different types of slit geometry: A V-shaped slit and a parallel slit. The parallel slit offers a greater exposure area to the beam; however, it results in a longer pulse duration, which worsens the energy resolution. Two different source sizes were tested to evaluate the intensity and energy resolution of each slit type. One source diameter is equal to the slit width, and one equal to the slit height. The discretized Monte Carlo simulations were coupled with the velocity of the chopper to evaluate the time structure of the pulse to stochastically estimate the energy resolution and intensity for the different cases. A monodirectional, monoenergetic source was used for the pulse time-structure simulations. Figures 2 and 3 illustrate the time structure of the pulses for the source diameter equal to the slit width and slit height, respectively. Both sets of simulations assumed the same chopper angular velocity, as well as the same slit height and slit width for the parallel and V-shaped slits.

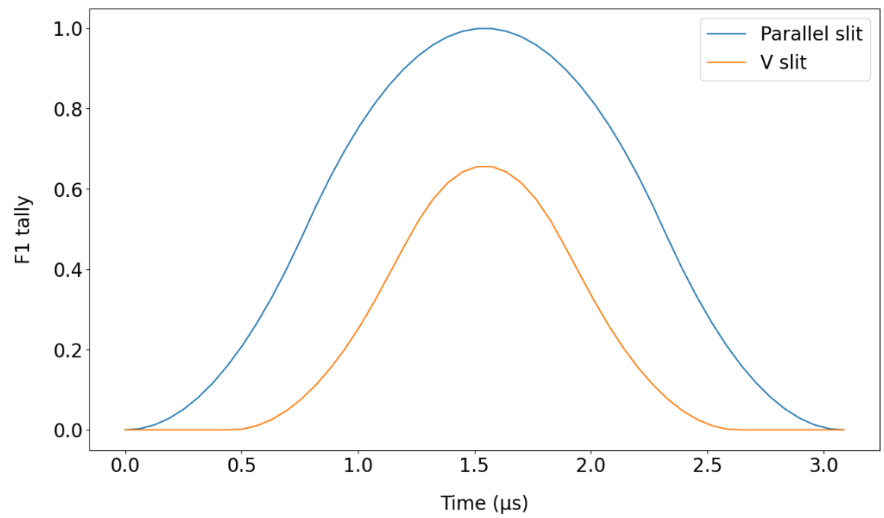

Figure 2. MCNP simulated time-structured pulse for the source diameter = 0.07 cm.

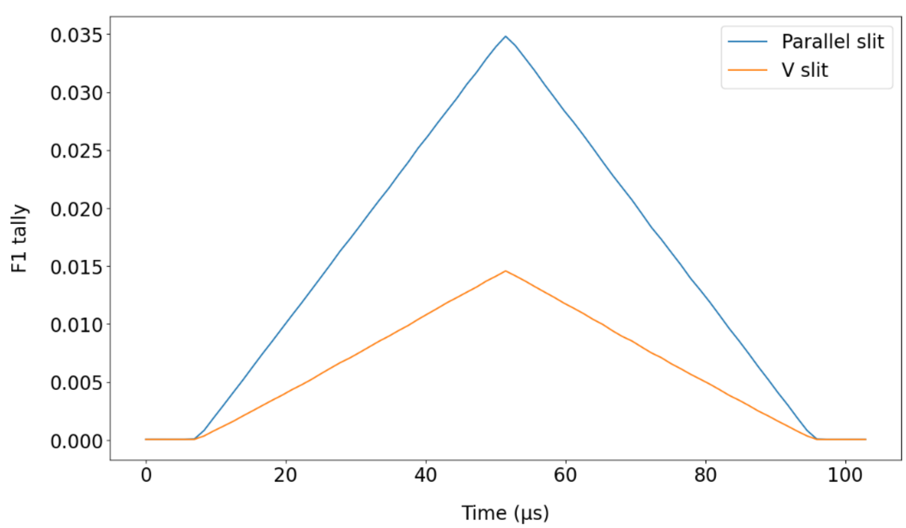

Figure 3. MCNP simulated time-structured pulse for the source diameter = 4 cm.

It is observed from Figures 2 and 3 that the duration of the pulse is drastically decreased when the source diameter is equal to the slit width. A source diameter larger than the slit width increases the pulse duration, while worsening the energy resolution. It is also observed that when the neutron source is larger than the slit, there is not full transmission of the source particles due to the partial blocking of the source. This is the reason for the sharp peaks shown in Figure 3. Tables 1 and 2 show the pulse structure information obtained from the MCNP6.2 simulations for the two source sizes for parallel and V-shaped slits.

Table 1. Pulse structure for nominal neutron energy of 40 eV and source diameter = 0.07 cm.

Slit Type	FWHM (μs)	Pulse Duration (μs)	Energy Resolution (%)	Energy Resolution at 70% Angular speed (%)
V-shaped	0.44	0.95	1.38	1.97
Parallel	0.82	1.50	2.11	3.02

Table 2. Pulse structure for nominal neutron energy of 40 eV and source diameter = 4 cm.

Slit Type	FWHM (μs)	Pulse Duration (μs)	Energy Resolution (%)	Energy Resolution at 70% Angular speed (%)
V-shaped	50.6	55.4	80.8	115.5
Parallel	50.6	51.2	74.7	106.7

It is observed that in order to achieve the desirable energy resolution, the source diameter must not be larger than the slit width for a given set of system parameters. It should also be noted that while the V-shaped slit achieves a better energy resolution, it also reduces the neutron pulse intensity by a factor of two based on the analytical calculations. The Monte Carlo simulations also demonstrate a reduced pulse intensity for the V-shaped slit for a given slit height and width compared to the parallel slit. For the case where the source diameter is equal to the slit height, it is observed that pulse duration is shorter for the parallel slit compared to the V-shaped slit. This is likely due to how the tool defines the full width as the time between instances where the tallies are 1% of the peak tally.

<u>Reactor Spectrum Transmission</u>

The second set of simulations used the same discretized movement for the mechanical chopper, however, the full simulated reactor spectrum was used as the neutron source. These simulations demonstrate the component of neutrons transmitted while the chopper is in the closed, open, or partially open positions. A transmission of 1E-6 has been defined as acceptable for 40 eV neutrons. Figure 4 shows the results from these discretized simulations assuming the same starting time for each starting particle for independent simulations.

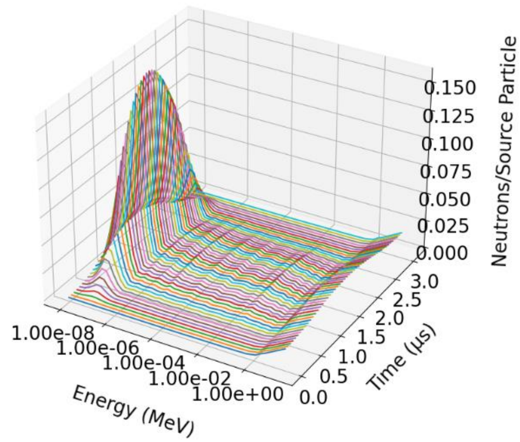

Figure 4. Discretized full spectrum transmission simulations with source diameter = 0.07 cm.

Figure 4 shows that while the chopper is in the closed position, neutrons of 40 eV and below are effectively blocked, and fast neutrons are transmitted. As the chopper transitions from closed to fully open, the intensity of neutrons below 40 eV drastically increases and decreases over the pulse duration for this set of system parameters. This illustration visually demonstrates how the chopper creates a pulse structed in both time and energy of neutrons transmitted.

Energy and Time Convolution

Simulations were also done to evaluate the TOF and the associated energy from neutron events that reach the target. These discretized simulations are useful for assessing events with the same TOF, but with differing energies to better understand the signal to noise ratios. Source particle start times were discretized to match the kinematics of the chopper, and tallies were binned into both energy and time. A running total for these tallies was created to evaluate the intensity of each bin for a full pulse. Figure 5 shows a heatmap created from these simulations where a grid of energy and time bins was created, and the summed tallies for each bin were mapped for intensity. It should be noted that the target is assumed to be 12 meters from the source.

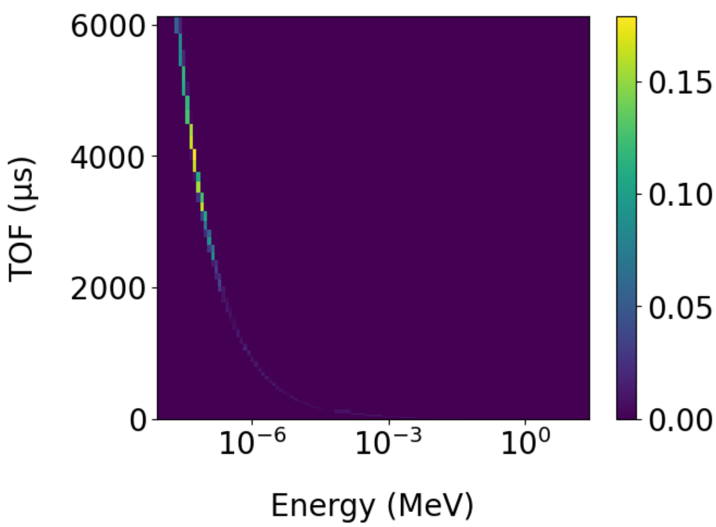

Figure 5. Heatmap of energy and time convoluted tallies for discretized pulse simulations.

Theses simulations are useful in evaluating neutron TOF for transmission analyses, while observing the intensity of specific energies that reach the target at specific times. This information can be used to correlate events to prompt gammas that will be measured in situ. These simulations can also be used to quantify the severity of frame overlap from subsequent pulses and uncertainty in neutron measurements.

ACTIVITY ESTIMATION

The Monte Carlo simulations discussed will serve as an estimation of the pulsed neutron source for activation predictions. Activity predictions will be used to design experiments for different samples. An activity prediction tool has been created to optimize experimental parameters for a given target. The optimization considers factors such as an allowable power level for the beamport in the unplugged configuration, gamma and beta dose from the sample, decay correction factors, detection efficiency and counting statistics, as well as argon-41 production in the beamport. Figure 6 shows an experimental parameter data set for a gold foil irradiation using the unfiltered beamport 1 of the PSBR. The source of neutrons was assumed to be the previously simulated spectrum binned into thermal, epithermal, and fast groups.

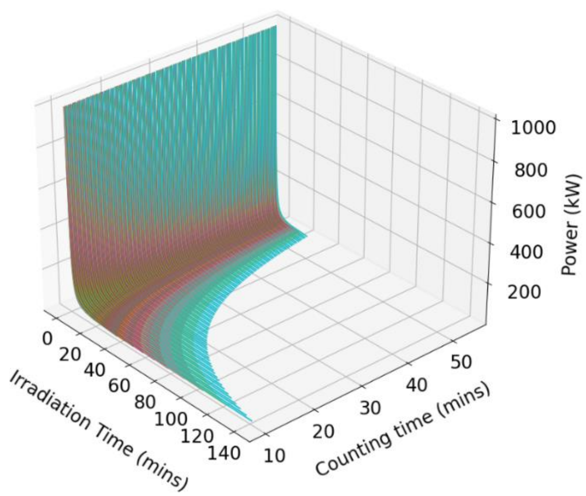

Figure 6. Experimental parameter data set for a gold foil irradiation.

This experimental data set created by the optimization tool is used to set the experimental parameters. Using this tool for a given target, the optimal power level, irradiation time, counting time and cooling time can be determined. The Monte Carlo simulation tool to evaluate the neutronics performance of the mechanical chopper will be coupled with the activation tool to inform experimental parameters for interrogating samples of interest.

CONCLUSIONS

A Monte Carlo tool has been created to validate analytical calculations for the mechanical neutron chopper at the PSBR. Analytical calculations have been used to optimize the chopper geometry, velocity and neutron intensity. These system parameters were simulated with MCNP6.2. Two types of slit geometry and source sizes were compared for their resulting energy resolution and intensity. The parallel slit offers a greater neutron intensity, while producing worse energy resolution. Deterministic and stochastic results for the energy resolution are in good agreement. The effect of increasing the neutron source size to be larger than the slit width degrades the energy resolution. A full spectrum, time-dependent simulation was performed to demonstrate the energy dependent transmission during the chopper operation. Energy and time convoluted simulations were created to evaluate the dynamic nature of the chopper and its effect on the neutron TOF. This can be used to quantify characteristics such as frame overlap or measurement uncertainties. Results from the Monte Carlo simulations will be used in conjunction with the activity prediction tool to design and optimize all relevant experiment parameters.

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