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An Evaluation of Systematic Biases of the Point Kinetics Model for Active Interrogation of Uranium[1]

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Abstract - *The point kinetics model relies upon a series of assumptions that simplify the process of neutron fission chain propagation. Tagged neutron interrogation of uranium using 14.1 MeV neutrons can potentially benefit from the use of the point kinetics model, but the appropriateness of the model's assumptions has yet to be validated for this application. The two assumptions considered by this work are that a single distribution can be used to describe the neutron multiplicity of a chain-starting event and that parasitic neutron capture within uranium is negligible. Monte Carlo simulations are used to evaluate the appropriateness of each assumption, as well as the consequences on the emitted doubles rate. This work demonstrates that the use of the point kinetics model for this application is possible, but corrections to the source multiplicity assumption are necessary that would likely require prior knowledge of sample isotopics.*

I. INTRODUCTION

The point kinetics model has historically been used as a framework to estimate characteristics of fissile material samples from coincidence and multiplicity measurements. Characteristics of interest to nuclear safeguards and nuclear security applications include fissile mass, total and leakage neutron self-multiplication, and isotopic ratios.

The point kinetics model has recently been explored as a method of estimating self-multiplication and enrichment of uranium metal under tagged neutron interrogation (TNI) measurements [1]. These measurements perform transmission imaging as well as fission site imaging, providing geometric and material identification information that can be used to constrain the solution space when inverting the point kinetics model equations to solve for neutron multiplication or uranium enrichment [2]. The point kinetics model is advantageous in that it allows instantaneous estimation of relevant characteristics when the number of observables is limited, as often is the case in coincidence and multiplicity counting. This simplicity comes at a cost, however. In order to simplify the neutron multiplication process, the point kinetics model makes several limiting assumptions about the internal process of fission chain propagation and the external process of detecting emitted neutrons. These assumptions may result in systematic biases that limit the usefulness of the point kinetics model to a small subset of applications, and these systematic biases must be evaluated before applying the model to a new application.

This work analyzes effects of the point kinetics model assumptions that concern how neutron fission chains begin and are propagated within a uranium sample. Specifically, it examines the assumption that all fission chains are initiated by a 14.1 MeV induced fission on ^{235}U or ^{238}U and the assumption that parasitic capture of fission neutrons is negligible. Monte Carlo simulations were used to evaluate the appropriateness of these assumptions. Data from the Monte Carlo simulations were also used to evaluate the consequences of each assumption by comparing the emitted neutron doubles rate as calculated by the point kinetics model equations under each assumption to a Monte Carlo estimate.

II. THEORY

1. Source Interaction Assumption

The point kinetics model assumes separate interactions for chain-starting interactions (0th generation) and induced neutron interactions (1st or greater generation) [3]. The definition of these events determines the values for the source and induced event multiplicity moments in the model equations, ν_{sn} and ν_{in}, respectively. When the point kinetics model is applied to the passive assay of plutonium, the chain-starting or source event is assumed to be the spontaneous fission of ^{240}Pu, with corrections for the contributions of α,n interactions. The induced event is assumed to be 2 MeV induced fission on ^{239}Pu [4]. The energy of 2 MeV is chosen because this is the average energy of neutrons emitted by induced fission on ^{239}Pu.

Common applications of the point kinetics model for active assay of uranium assume that neutron fission chains are initiated by source neutrons inducing fission on ^{235}U. The assumption is typically justified for systems such as the Active Well Coincidence Counter, which uses an americium-lithium (AmLi) neutron source [5]. The AmLi source produces neutrons with an average energy of 0.3 MeV. Most of the neutrons produced by the AmLi source are of energies below the region where ^{238}U, the most abundant isotope in most uranium samples, has a significant induced fission cross section. This assumption is not as appropriate when using a deuterium-tritium (D-T) generator as the interrogating source. As shown by Table I, the ^{238}U induced fission cross section cannot be

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ignored for 14.1 MeV source neutrons. The cross sections for n,2n and n,3n interactions, which are indistinguishable from fission in TNI measurements, are also significant compared to the fission cross sections, particularly for ^{238}U.

TABLE I. Uranium Microscopic Cross Sections for 14.1 MeV Neutrons (barns) [6]

	σ_{nf}	σ_{n2n}	σ_{n3n}
^{235}U	2.080	0.522	0.033
^{238}U	1.143	0.881	0.406

The point kinetics model assumption that all neutron fission chains are initiated by 14.1 MeV neutrons inducing fission on either ^{235}U or ^{238}U manifests itself in the point kinetics model equations by affecting the ν_{sn} values. The multiplicity distribution, and consequently the ν_{sn} values, for 14.1 MeV induced fission on ^{235}U is estimated using the Terrel method [7], with the $\bar{\nu}$ taken from ENDF/B-VII.1 [6]. Previous work suggests that the multiplicity distribution for 14.1 MeV induced fission on ^{238}U is similar to that of ^{235}U [8]. Values for the ν_{in} values are calculated from the multiplicity distribution for 2 MeV induced fission on ^{235}U measured by Zucker and Holden [9]. Induced neutron interactions occur at fission neutron energies. At these energies, the multiplicity distribution is largely insensitive to uranium enrichment.

Because n,γ, n,2n and n,3n interactions are indistinguishable from a fission with a multiplicity of zero, two, or three, respectively, these interactions can be considered when determining values for ν_{s1} and ν_{s2}. If a uranium sample's isotopics are known a priori, values for ν_{sn} can be estimated using the following equations:

$$\nu_{sn} = \sum_{\nu=0}^{N_{ind}} \frac{\nu!}{n!(\nu-n)!} \left(\frac{\Sigma_{nf} P_\nu + \Sigma_{n\gamma}\delta_{\nu 0} + \Sigma_{n2n}\delta_{\nu 2} + \Sigma_{n3n}\delta_{\nu 3}}{\Sigma_f + \Sigma_{n\gamma} + \Sigma_{n2n} + \Sigma_{n3n}} \right), \tag{1}$$

$$\Sigma_i = N_A \rho \sum_A \frac{f_A \sigma_i^A}{A}, \tag{2}$$

where N_{ind} is the highest value of neutron multiplicity ν observed, Σ_i is the macroscopic cross section for a source neutron undergoing interaction i, and P_ν is the probability that a source neutron-induced fission has multiplicity ν. The parameter f_A is the fraction of isotope A in the uranium, while σ_i^A is the microscopic cross section for a source neutron undergoing interaction i on isotope A. The mass density is represented by ρ and N_A is Avogadro's number.

2. Negligible Parasitic Capture Assumption

The point kinetics model combines the spatial, energy, isotopic, and directional variations in fission chain propagation into the single concept of average neutron multiplication. The increase in neutron population due to a single neutron is known as the total self-multiplication factor M_T. Another quantity of interest is the number of induced fissions per neutron, $p_f M_T$, where p_f is the probability that an induced fission neutron induces further fission. However, a description of the neutron population that escapes from a fissionable sample, and can thus be detected by an external detector, is necessary when applying the point kinetics model equations to a measurement scenario. In order to account for the neutron population increase due to fission and neutron population decrease due to parasitic capture, the leakage self-multiplication term M_L is often used. It is defined as the increase in the emitted neutron population as a result of one additional neutron in the system. It can also be defined as the ratio of neutrons leaking out of a fissionable sample to the number of neutrons created by the chain-starting event.

$$M_L = (1 - p_f - p_c) M_T = \frac{(1 - p_f - p_c)}{1 - p_f \nu_{i1}}, \tag{3}$$

where ν_{i1} is the average number of neutrons produced by an induced fission and p_c is the probability that a neutron is parasitically captured.

In order to limit the dimensionality of the point kinetics model equations, it is commonly assumed that parasitic capture of neutrons within the fissionable object is negligible. In other words, the only neutron losses within the sample are from inducing fission. This allows for leakage self-multiplication M_L to remain a function of only one characteristic parameter, p_f. This assumption manifests itself when the $p_f M_T$ term, which appears in the point kinetics model equations of order ≥ 2, is expressed in terms of only M_L. The following assumption is used [10]:

$$p_f M_T \approx \frac{M_L - 1}{\nu_{i1} - 1} = M_T \left[p_f - \frac{p_c / p_f}{\nu_{i1} - 1} \right]. \tag{4}$$

This approximation holds when $p_c / p_f \ll 1$.

III. DESCRIPTION OF THE ACTUAL WORK

A series of Monte Carlo simulations using MCNPX-PoliMi [11] were developed to evaluate the effect of these assumptions. In the simulations, 14.1 MeV neutrons are incident upon a uranium object of varying mass and enrichment. All the results shown in this paper are for a spherical geometry. A combination of PoliMi-specific outputs and surface-source-write outputs were parsed using custom scripts to calculate Monte Carlo estimates of several parameters relevant to the point kinetics model equations. Parameters include the emitted neutron multiplicity distribution, the multiplicity distribution for interactions by source (0th generation) and induced (1st or greater generation) neutrons, leakage and total self-multiplication, among many others.

Both of the assumptions addressed in this work are evaluated individually. The effects of each assumption are quantified by comparing the Monte Carlo estimate of emitted doubles rates to those calculated with the point kinetics model equation when operating under that assumption. The point kinetics model equation for the emitted doubles rate of a uranium object under TNI is

$$\Phi_2 = \beta M_L^2 \left[\nu_{s2} + \nu_{i2} \nu_{s1} p_f M_T \right], \tag{5}$$

where β is the probability that a source neutron (a 14.1 MeV neutron from a D-T neutron generator) initiates a fission chain.

The quantities ν_{i1} and ν_{i2} are the first and second reduced factorial moments of the prompt neutron multiplicity distribution for an induced neutron event, respectively. Similarly, ν_{s1} and ν_{s2} are the first and second reduced factorial moments of the chain-starting event prompt neutron multiplicity distribution, respectively. If the negligible parasitic capture assumption is applied, the approximation in Eq. (4) can be applied to Eq. (5):

$$\Phi_2 = \beta M_L^2 \left[\nu_{s2} + \nu_{i2}\nu_{s1} \left(\frac{M_L - 1}{\nu_{i1} - 1} \right) \right]. \quad (6)$$

The doubles rate equation in Eq. (6) is similar to the equation traditionally used in passive assay of plutonium [4]. The only differences are the coupling term β normalizes the rate to the number of source neutrons, and the ν_{s1} and ν_{s2} terms encapsulate all chain-starting interactions as opposed to only spontaneous fission.

The Monte Carlo estimate for the emitted doubles rate is compared to the doubles rate as calculated by different point kinetics model equations for the following cases:

0. Equation (5) using Monte Carlo estimates for all parameters. This serves as a sanity check to ensure the point kinetics model equation and faithfully represents the Monte Carlo simulations.

1. Equation (5) using Monte Carlo estimates for M_L, p_f, and M_T. The values for ν_{s1} and ν_{s2} are based on the assumption that all fission chains are initiated by 14.1 MeV neutron-inducing fission on either ^{235}U or ^{238}U. This evaluates the effect of the source interaction assumption while controlling for the effect of the negligible parasitic capture assumption.

2. Equation (5) using Monte Carlo estimates for M_L, p_f, and M_T. Estimates for ν_{s1} and ν_{s2} are calculated using Eqs. (1) and (2), assuming enrichment is known a priori. This evaluates the effect of taking additional source interactions into account while controlling for the effect of the negligible parasitic capture assumption.

3. Equation (6) using Monte Carlo estimates for M_L, ν_{s1} and ν_{s2}. This evaluates the effect of the negligible parasitic capture assumption while controlling for the effect of source interaction assumption.

4. Equation (6) using the Monte Carlo estimate for M_L. The values for ν_{s1} and ν_{s2} are based on the assumption that all fission chains are initiated by 14.1 MeV neutron-inducing fission on either ^{235}U or ^{238}U. This evaluates the effect of applying both assumptions.

The Monte Carlo estimate for the coupling coefficient β is used in the point kinetics model equations for all of the above cases.

In addition to these cases, the validity of the $p_c/p_f \ll 1$ assumption is also evaluated by taking the ratio of the Monte Carlo estimates of these probabilities. Because this assumption was initially made for and is used widely in passive assay of plutonium, a Monte Carlo simulation of a sphere of weapons-grade plutonium was also performed to estimate the p_c/p_f ratio compared to the uranium results. The physical characteristics of the plutonium were taken from Ref. [12]. In all simulations, an adequate number of Monte Carlo trials were run to ensure that the statistical uncertainty in the estimated parameters, as well as the calculated values for the emitted doubles rates in Eqs. (5) and (6), the percent errors in doubles rate estimation, and the p_c/p_f ratio, was less than 0.1 percent. The uncertainty in these calculated values is calculated by propagating the Monte Carlo statistical uncertainties of each parameter in the respective equation.

IV. RESULTS AND ANALYSIS

The results for Case 0, which serves as a sanity check and verification of the methods, are shown in Fig. 1. The point kinetics model equation for the doubles rate as expressed in Eq. (5) matches the Monte Carlo estimates for the emitted doubles rate to within one percent error when Monte Carlo estimates for all parameters are used. This implies that any observed differences for the remaining cases can be attributed to the assumptions made by that case, to within one percent.

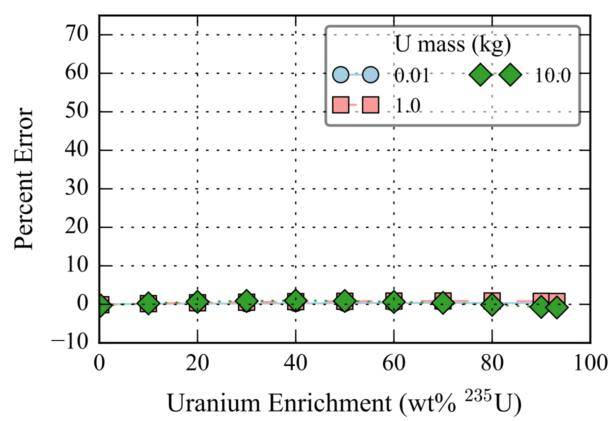

Fig. 1. Case 0 results. Percent error between the Monte Carlo doubles estimates and doubles estimates using the point kinetics model equation Eq. (5) and Monte Carlo estimated values for all parameters. The statistical uncertainty in the percent error due to propagated Monte Carlo uncertainties is negligible on the shown scale.

1. Source Interaction Assumption

The point kinetics model assumption that all neutron fission chains are initiated by 14.1 MeV neutrons inducing fission on ^{235}U or ^{238}U (Case 1) results in an overestimation of the emitted doubles rate, as shown in Fig. 2. This overestimation is a result of ignoring the n,2n and n,3n interactions by 14.1 MeV neutrons on both ^{235}U and ^{238}U, which drives down the average number of neutrons and neutron doubles from a source event. Because ^{238}U has a comparatively larger cross section for these interactions, lower enrichment simulations exhibit a much larger overestimation by the point kinetics model equation when these interactions are ignored.

Consequently, there is significant improvement in the

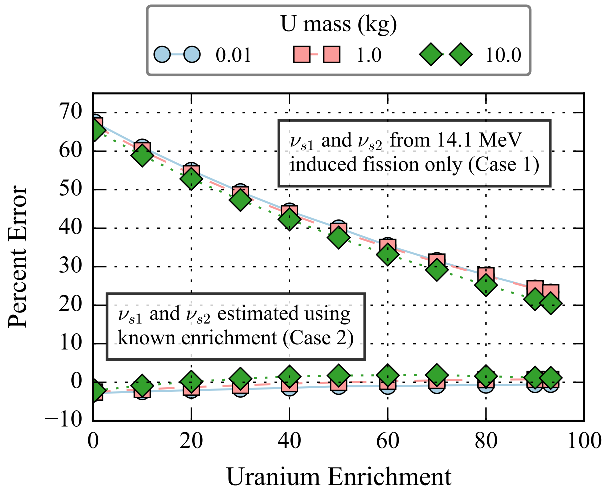

Fig. 2. Case 1 and 2 results. Comparison of the percent error between the Monte Carlo doubles estimate and the point kinetics doubles estimates for each case. The statistical uncertainty in the percent error due to propagated Monte Carlo uncertainties is negligible on the shown scale.

doubles rate estimate when a priori knowledge of sample enrichment is applied to Eqs. (1) and (2) to estimate ν_{s1} and ν_{s2} (Case 2), as shown in Fig. 2. The degree to which n,2n and n,3n interactions ultimately influence ν_{s1} and ν_{s2} values is enrichment dependent due to the higher cross sections for these interactions on ^{238}U versus ^{235}U. Even for simulations of uranium with greater than 93% ^{235}U, however, there is at least 20 percent overestimation versus Monte Carlo estimates for emitted doubles rates. This demonstrates that accounting for all neutron-producing interactions when determining chain-starting multiplicity constants can significantly improve the accuracy of the point kinetics model equations for TNI of bare uranium metal of any enrichment. The downside to this is that isotopic information must be known or estimated a priori. However, this analysis suggests that just simply assuming all chain-starting interactions to be 14.1 MeV induced fission on ^{235}U or ^{238}U, thereby eliminating the source of isotopic sensitivity, injects too much systematic bias into the point kinetics model equations for them to be useful.

2. Negligible Parasitic Capture Assumption

Figure 3 shows the assumption condition $p_c/p_f \ll 1$ to be considerably worse compared to the passive plutonium simulation. The largest p_c/p_f ratios were seen in simulations with greater concentrations of ^{238}U. This is largely explained by the difference in the macroscopic cross section ratios $\Sigma_{n\gamma}/\Sigma_{nf}$ between ^{238}U and ^{235}U. Using fission energy-averaged cross sections, this ratio is approximately 0.237 and 0.0785 for ^{238}U and ^{235}U, respectively [13]. However, the p_c/p_f ratios for the 0.3 percent ^{235}U simulations increased well beyond the ^{238}U $\Sigma_{n\gamma}/\Sigma_{nf}$ ratio, with p_c/p_f increasing with total uranium mass. This is likely a result of a downscattering effect from inelastic scatter in ^{238}U. Neutrons born inside larger objects will undergo more scatters and lose more energy before removal

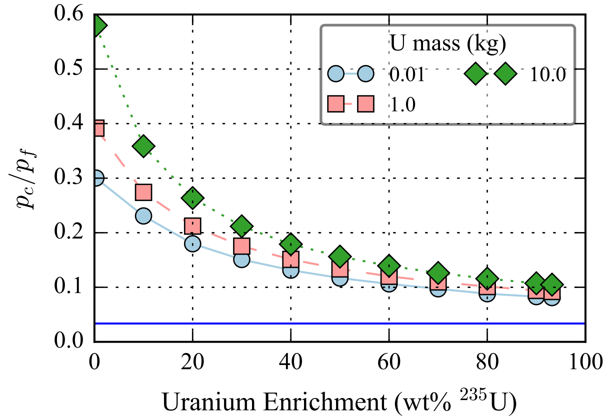

Fig. 3. Monte Carlo estimates of the induced neutron capture-to-fission ratio for uranium. The solid blue line represents the Monte Carlo estimate for a passive plutonium simulation. The statistical uncertainty in the p_c/p_f ratios due to propagated Monte Carlo uncertainties is negligible on the shown scale.

compared to those born inside smaller objects, as shown in Fig. 4.

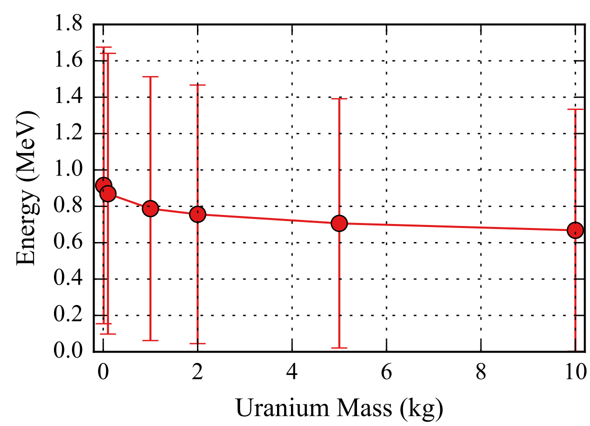

Fig. 4. Energy of induced neutrons (generation ≥ 1) when undergoing a removal interaction (fission, capture, nxn) versus total uranium mass for 50% ^{235}U case. Error bars are one standard deviation of the energy distribution as estimated by Monte Carlo. These distributions are highly non-Gaussian.

Despite the significant violation of the point kinetics model assumption that parasitic capture within the fissionable object is negligible for all uranium simulations, this violation results in only a very slight underestimation of the emitted doubles rate (Case 3), as shown in Fig. 5.

This insensitivity of the doubles rate to the p_c/p_f ratios can be understood in the context of the point kinetics model for uranium. Any bias in the doubles rate due to assuming negligible parasitic capture only affects the second term in Eq. (5), which represents the doubles due to fission chains initiated by neutrons from the chain-starting reaction. The magnitude of this contribution to the doubles rate varies with uranium enrichment. At lower enrichment, the magnitude of this contribution is low. Self-multiplication is quite low, typically near unity, and most doubles come from the initial chain-starting reaction. Thus the ultimate effect of a large p_c/p_f ratio on the doubles rate is small. At higher enrichment, the contribution to the doubles rate is comparatively greater. However, the p_c/p_f ratios are lower, so the doubles rate remains insensitive to the effect of parasitic capture.

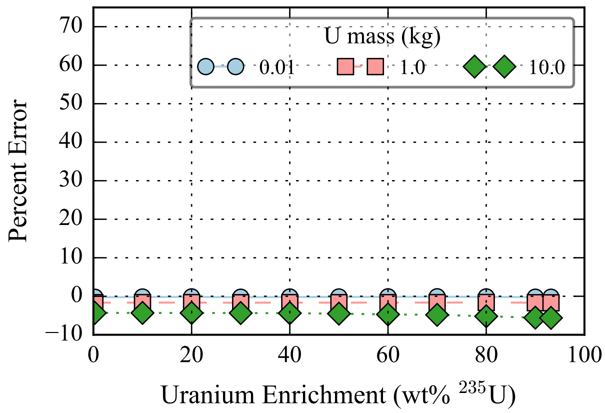

Fig. 5. Case 3 results. Percent error between the Monte Carlo doubles estimates and doubles estimates when only the negligible parasitic capture assumption is applied. The statistical uncertainty in the percent error due to propagated Monte Carlo uncertainties is negligible on the shown scale.

3. Both Assumptions Simultaneously

Figure 6 shows the results for Case 4, where both the assumption that all neutron fission chains are initiated by a single interaction, namely 14.1 MeV neutrons inducing fission on ^{235}U or ^{238}U, and the assumption that parasitic neutron capture is negligible are considered. These results are only slightly different from the results for the source interaction assumption by itself (Case 1) shown in Fig. 2. This is to be expected considering the insensitivity of the doubles rate to parasitic capture. The competing effects of these two assumptions result in a slightly lower overestimation of the Monte Carlo estimated doubles rate compared to Case 1. Since the source interaction assumption affects the multiplicity of fission chain generation 0, while the negligible parasitic capture assumption affects the emitted multiplicity for generations ≥ 1, it is appropriate to assume that these two effects are uncorrelated. This is further substantiated by the observation that the Case 4 percent errors are nearly a perfect additive combination of the percent errors of Case 1 and Case 3, implying that the covariance in the sensitivity of these two effects is negligible. The magnitude of both of these effects appears of be sensitive to physical parameters such as mass and enrichment to varying degrees, so there may be some small covariance between them. However, these results indicate that it is not significant enough to warrant any corrections within the point kinetics model.

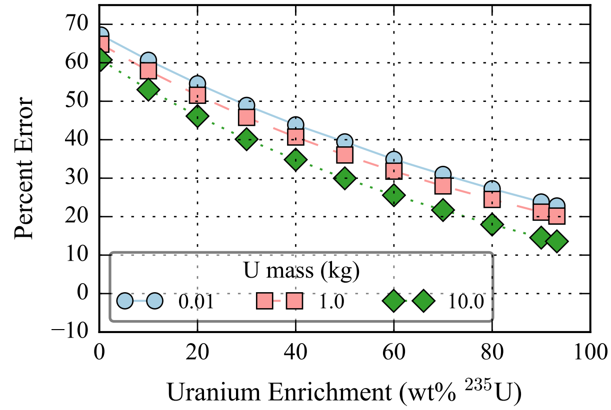

Fig. 6. Case 4 results. The effect of assuming all source interactions are induced fissions by 14.1 MeV neutrons on ^{235}U or ^{238}U and negligible parasitic capture. The statistical uncertainty in the percent error due to propagated Monte Carlo uncertainties is negligible on the shown scale.

V. CONCLUSIONS

This work examined two sources of systematic bias in the point kinetics model equations developed for active interrogation of bare uranium metal by 14.1 MeV neutrons. Results determined that at 14.1 MeV, other neutron-producing reactions that cannot be distinguished from induced fission can significantly bias the estimation of neutron doubles rate by the point kinetics model equations if they are not accounted for. The magnitude of this effect is sensitive to uranium enrichment, so knowledge of sample isotopic information may be necessary to make adequate corrections. On the other hand, the assumption that parasitic neutron capture is negligible, while shown to be violated, is also shown to introduce significantly less bias and is largely uncorrelated to the bias introduced by assuming that all neutron fission chains are initiated only by 14.1 MeV induced fission on ^{235}U or ^{238}U.

If the point kinetics model is to be utilized by TNI measurements to estimate physical parameters of interest from coincidence and multiplicity measurements, the systematic biases inherent in the model must first be quantified and understood. This work suggests that using isotopic information to estimate the chain-starting multiplicity may be necessary to produce accurate results. Future research should examine scenarios where isotopic information cannot be assumed a priori and explore alternative formulations of the point kinetics model equations that may be able to reduce or correct for this enrichment sensitivity. The imaging capabilities of TNI methods may also offer additional information that could be used to address this enrichment-dependent systematic bias.

VI. ACKNOWLEDGMENTS

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