Neutron Noise Analysis Techniques for Improved Verification of Critical and Subcritical Cores

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Safeguards verifications at research reactors and (sub)critical assemblies are often challenged by the limited access to or complete inaccessibility of in-core material. Additional difficulties are related to the significant impact of reactor design and its operation history on the emitted radiation. In recent years, the IAEA has expanded a standard toolkit of its in-core material verification techniques, which were traditionally based on total neutron counting, towards application of advanced approaches involving detection of correlated neutrons and Monte Carlo modelling. These new verification techniques improved the confidence level for verification of direct use nuclear materials, while reducing the burden on the facility operator.

In this paper we present further advancements of the verification techniques, with particular emphasis on the Feynman-α and power spectral density (PSD) neutron noise analyses and their application in quantitative verification of the fast highly enriched uranium core of the TAPIRO research reactor (ENEA, Casaccia Research Centre, Italy). In the course of this research and development effort, multiple neutron noise measurements were performed at different reactor criticalities (ranging from deep subcritical to supercritical states) and reactor power levels. The data were acquired using a pulse digitizer capable of recording long waveforms from a shielded 1 in. $\times$ 1 in. stilbene detector positioned near the reactor core. To accurately process and interpret the data thus obtained, a new Monte Carlo-based approach involving a multi-group representation of the prompt neutron generation time distribution was developed and utilized. The multi-group approach overcomes shortcomings of the classical single-group point-kinetics model observed when applied to fast reflected cores, thereby providing more accurate estimates of the correlation magnitude (Y_∞) and temporal scale (α) of the neutron multiplication process. A selected combination of measurable characteristics, including the α value and a newly introduced λ-parameter, provided a robust approach for the verification of both reactor design and in-core fissile mass. The λ-parameter is independent of the reactor criticality and power levels, yet highly sensitive to removal of small quantities of nuclear material from the core and modifications of the reactor and core designs.

1. Introduction

Safeguards verifications at research reactors, critical assemblies and subcritical cores are often hampered by the partial or complete inaccessibility of in-core material. Additional difficulties are imposed by the peculiarities and design features of particular nuclear installations, their operational status and history, radiation conditions, as well as by the availability of experimental channels and appropriate locations for performing verification measurements. Of no less importance are factors such as intrusiveness and the operational burden that may be associated with safeguards verification activities and should be minimized as much as possible.

The criticality check, a standard IAEA verification technique [1], has been routinely utilized for the verification of research reactors and critical assemblies. The technique is based on the confirmation of a supercritical state of a reactor upon insertion of a small positive reactivity into a verified core. Supercriticality is confirmed by the exponential growth in the measured total neutron counts. Recently, the technique has been extended to include the rod drop test and analysis of the exponential decay of the total neutron counts after reactor scram.

As the applicability of the criticality check is limited to critical systems, a verification technique based on the determination of reactivity ratios at several subcritical states was suggested in Ref. [2] for situations involving subcritical assemblies and non-operational or shutdown cores. It was demonstrated that combining the reactivity ratios technique with Monte Carlo simulations would allow partial defect verification of in-core material.

By providing direct evidence of the fissile material presence in a verified core, neutron correlation measurements bring additional advantages in terms of higher robustness and confidence, compared to total neutron counting. A successful deployment of the neutron correlation-based verification approach, including neutron noise measurements and extended Monte Carlo computations, for quantitative verification of liquid fuel in a decommissioned research reactor was reported in Ref. [3].

In this paper we present further advancements in the in-core verification techniques, with emphasis on the application of the Feynman-α and PSD neutron noise analysis methods for the verification of fast cores. The proposed innovations are demonstrated in the example of the TAPIRO research reactor, operated by the Italian National Agency for New Technologies, Energy and Sustainable Economic Development (ENEA) at the Casaccia Research Centre near Rome.

2. Theoretical

Reactor noise analysis is one of the basic reactor diagnostics methods [4]; it makes use of the fundamental property of multiplying media appearing as an excess variation in neutron emissions above the pure random (Poisson) variance. According to the single-group point-kinetics theory [5], the excess variance-to-mean ratio for the number of neutrons $N(t)$ detected by an instantaneous point detector within a time interval of length t is given by the following formula:

$$Y(t) = \frac{\sigma_N^2(t)}{\langle N(t) \rangle} - 1 = Y_\infty \times \left(1 - \frac{1 - \exp\left(-\alpha t\right)}{\alpha t}\right) \qquad \text{Eq. 1}$$

where Y_∞ and α are characteristic measures of the neutron correlation and temporal evolution of the multiplication process within fission chains, respectively. Both quantities can be expressed as functions of the reactor kinetic and detector parameters:

$$Y_\infty = \frac{\varepsilon\, D_\nu}{(\beta_{eff} - \rho)^2}, \quad \alpha = \frac{\beta_{eff} - \rho}{\Lambda}, \quad \rho = \frac{k_{eff} - 1}{k_{eff}}, \quad D_\nu = \frac{\langle \nu(\nu - 1) \rangle}{\langle \nu \rangle^2} \qquad \text{Eq. 2}$$

where ε is the detection efficiency, β_{eff} is the effective delayed neutron fraction, Λ is the prompt neutron generation time, ρ is the delayed reactivity, k_{eff} is the effective neutron multiplication factor, and D_ν is the Diven factor, defined as the ratio of the second and first factorial moments of the fission neutron multiplicity distribution $P(\nu)$ [6].

Despite the possibility demonstrated in [7] to establish the connection between the conventional passive neutron coincidence counting (PNCC [8]) and neutron noise analysis (NNA) techniques, some essential differences between them are worth emphasizing. Particularly, NNA is normally applied to massive, close-to-critical, highly multiplying objects such as reactor cores, whereas PNCC usually deals with relatively small, deeply subcritical samples. The PNCC analysis is heavily based on the super-fission (i.e., instant fission) concept, whereas one of the essential fundamentals in the NNA is the detailed consideration of the temporal dynamics of neutron multiplication inside an examined object. Moreover, NNA benefits from the application of near-instantaneous, point-like detectors (i.e., super-detectors) imposing minimal distortion onto measured neutron pulse trains. By contrast, PNCC employs bulky moderating neutron counters that largely obfuscate unique temporal characteristics inherent in neutron emissions emerging from a measured sample.

3. Experimental

3.1. *TAPIRO reactor*

The TAPIRO zero power nuclear research reactor is a fast neutron source that has been in operation since 1971 [9]. The 12-cm-diameter cylindrical core is made of a dense alloy of highly enriched uranium (HEU) and molybdenum (18.5 g.cm^{-3}, 1.5 wt% Mo, 93.5 wt% ^{235}U). The reactor design includes a cylindrical copper reflector composed of inner and outer parts, with an external diameter of 80 cm and total weight of 2.6 tons. Being close to unity, the height-to-diameter ratio of both the core and reflector ensures low neutron leakage and good spherical symmetry of the neutron flux, with up to $4{\cdot}10^{12}$ cm^{-2}.s^{-1} of nearly pure fission spectrum neutrons in the core centre and variable spectrum hardness in adjacent locations. The design kinetic parameters of the core are: Λ = 50 ns, β_{eff} = 0.0066.

The availability of several radial and tangential experimental channels (see Figure 1) allowed the reactor to be utilized for numerous experimental programs on fast reactors, materials testing, metrology and basic nuclear research. For the investigations described herein, multiple neutron noise measurements were performed at different reactor power levels and different reactor criticality states ranging from deep subcritical to supercritical.

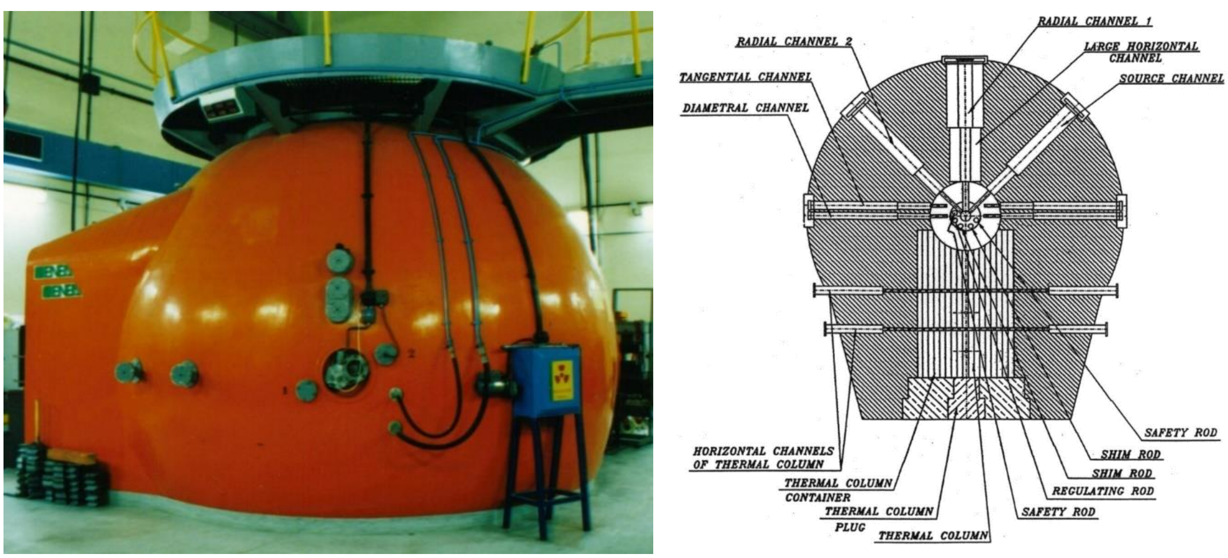

Figure 1. The TAPIRO research reactor (left) and its horizontal centre core cross section (right).

3.2. *Equipment*

The measurements were performed using the SD7750S integrated fast neutron data acquisition system from CAEN SpA [10], which included the R77505 14-bit flash analog-to-digital converter waveform digitizer, a 4 kV / 3 mA high-voltage power supply module and a high-performance industrial computer. The data acquisition system was coupled to a fast scintillation detector from Inrad Optics, embedding a 1″ × 1″ Scintinel™ stilbene single crystal and a Hamamatsu R6094 photo-multiplier tube. The detector waveforms were digitized at the 500 MS/s sampling rate (2 ns sampling interval) and transferred to the computer via an optical link at the 80 MB/s speed. The WaveDump application [11] was utilized to record bunches of randomly triggered 10.48-ms-long waveforms. Each waveform comprised over 5.24 million continuous samples from the detector output signal.

3.3. *Measurements*

The detector assembly was positioned inside the copper reflector, at the far end of radial channel 2. The distance from the front end of the stilbene crystal to the core centre was about 29 cm. To reduce the potential impact of the direct and scattered prompt/decay gamma rays, the detector was shielded by 20 mm and 40 mm thick layers of lead on its side and front, respectively.

Data were acquired throughout multiple runs in six subcritical states achieved via a stepwise increase of the safety rod positions, covering the range of the effective multiplication factor $k_{eff} = 0.9925 \div 0.9982$ and negative reactivity $\rho = -111$ ¢ ÷ -26 ¢. The measurements were performed with the Am-Be start-up neutron source (I ≈ 10^7 n.s^{-1}) positioned near the reactor core. During the critical and supercritical (transient) state measurements, the start-up source was brought to its "parked" position far away from the core. The critical state data were acquired at power levels of 1 mW and 10 mW. The transient state was initiated by inserting a positive reactivity of $\rho = +13$ ¢ at 1 mW power.

Each acquisition run took about 30 s, producing up to 250 individual waveforms for further offline analysis. Waveform examples are shown in Figure 2.

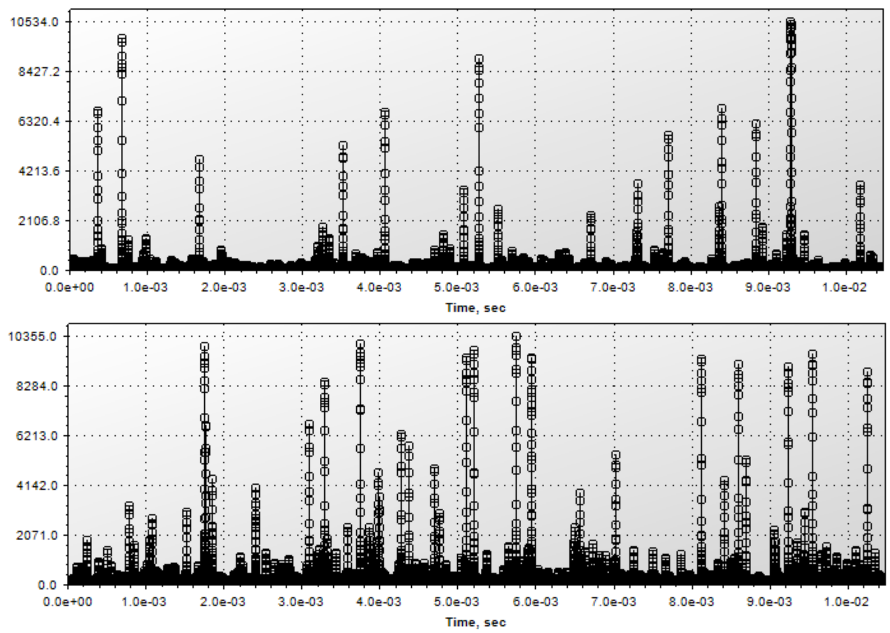

Figure 2. Waveforms acquired from a subcritical (k_{eff} = 0.9948, top) and critical (10 mW, bottom) reactor.

3.4. *Data processing*

The measured waveforms were processed to obtain estimates for the Y_∞ and α parameters. Two methods, Feynman-α and PSD, were independently employed for this purpose [12]. The Feynman-α analysis included extraction of pulse arrival times from the waveforms, creation of pulse trains and calculation and fitting of the resulting Feynman-Y data arrays. The PSD analysis required preconditioning (coarse graining and truncation) of the waveforms, followed by fast Fourier transform of the conditioned data, subtraction of the instrumentation noise and fitting of the resulting spectral data points.

Analytical representations of the fitting functions were obtained from Monte Carlo calculations of the prompt neutron generation time distribution and detector temporal response functions. The application of a multi-group model for the prompt neutron generation time distribution was essential for overcoming deficiencies inherent in classical neutron noise methods stemming from a common use of a single-group point-kinetics model, which is not applicable in the case of fast reflected cores.

4. Safeguards verification methodology

4.1. *Lambda parameter*

Eq. 2 suggests that the quantity $\lambda = Y_\infty^{1/2}\cdot\alpha$ does not depend on the reactor criticality level and will remain constant as long as the reactor design and detector positioning have not been changed:

$$\lambda = \sqrt{Y_\infty}\cdot\alpha = \frac{\sqrt{D_\nu\cdot\varepsilon}}{\Lambda} \qquad \text{Eq. 3}$$

A more generic definition of the λ parameter, allowing a deviation of the Y_∞ vs. α dependence from its theoretical form $Y_\infty \sim \alpha^{-2}$, can be introduced as follows:

$$\lambda_b = Y_\infty^{1/b}\cdot\alpha = \frac{(D_\nu\cdot\varepsilon)^{1/b}}{\Lambda} \qquad \text{Eq. 4}$$

In the case of the TAPIRO reactor, the experimentally determined coefficient $b \approx 1.8$ deviates slightly from its theoretical value $b = 2$.

Figure 3 shows the results of the neutron noise analysis, which are plotted as $Y_\infty^{1/b}$ vs. α^{-1}. In this special representation, the data points group along zero-offset straight lines with slopes defined as $\tan\varphi = \lambda_b$. Different slopes in the case of the Feynman-α and PSD data points are explained by the specifics of the data processing algorithms employed in both methods, e.g., pulse height discrimination, coarse graining, filtering etc. These peculiarities effectively result in different detection efficiencies in Eq. 3 and Eq. 4, hence, different λ_b values: = 0.298(3) for the Feynman-α and = 0.221(2) for the PSD analysis.

4.2. *Design verification*

Since both Y_∞ and α can be measured using one of the reactor noise analysis techniques, the experimentally determined λ value can be utilized to confirm that the reactor design has not been altered. The possibility to measure λ value at either a critical or subcritical state translates into less time and effort required to conduct the verification activities, as well as the possibility to confirm the design information of non-operational and subcritical cores.

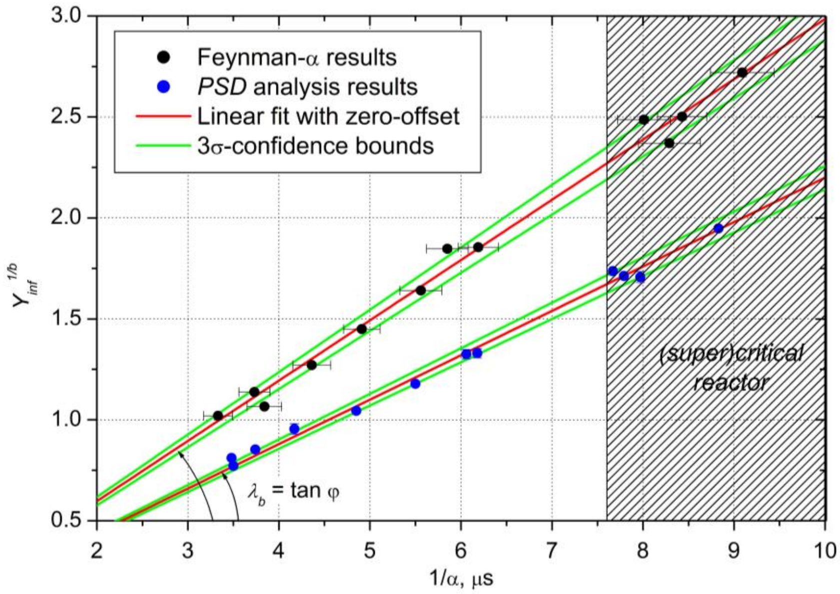

Figure 3. Results of the neutron noise measurements at the TAPIRO reactor. The error bars represent statistical uncertainties with coverage factor k=3. The shaded area highlights the domain of the reactor (super)criticality.

The high importance of design verification stems from the fact that different core and reflector designs may require substantially different amounts of nuclear material for reaching criticality. Therefore, implementing changes in the reactor design is considered to be one of the plausible pathways for concealing a potential diversion. To find out how effective the λ_b parameter is in identifying such design modifications, a sensitivity study was conducted using the TAPIRO reactor as an example.

The first design alteration scenario examined whether adding an efficient moderating material could enable criticality to be reached with a smaller amount of HEU. Apart from some exotic substances such as liquid or solid methane, high-density polyethylene (HDPE), a material with an extremely high hydrogen atomic density, is the top candidate in this scenario.

A series of MCNP simulations considered removals of certain fractions of the HEU from the core. For each fraction removed, increasing amounts of HDPE were added to (uniformly mixed with) the remaining HEU fuel until criticality was regained. As the core dimensions were changing, the height-to-diameter ratio was kept at its optimal value ($H/D = 0.8$), ensuring minimal neutron leakage. Once a new critical configuration had been determined, new Λ, D_ν, ε and λ_b values were calculated. The results of the calculations are presented in Table 1.

The data show that λ_b is extremely indicative of design modifications that involve the addition of moderating materials. Specifically, λ_b values drop by up to 74% when portions of HEU from 0% to 12% are gradually removed from the core. The high sensitivity of λ_b parameter to the design alterations considered in these simulations is a result of a significant increase in the prompt neutron generation time and a substantial loss of detection efficiency, both effects being caused by a considerable softening of the neutron spectrum due to thermalization.

Table 1. Results of the MCNP calculations for a hypothetical TAPIRO core with HDPE added to counterbalance the reactivity loss due to removal of various δm_{235} fraction of HEU from the core. The calculated Λ and λ_b values are shown as ratios to their values Λ_0 and λ_{b0} at the nominal core design. Detector efficiency was calculated assuming the low-level threshold of 44 keV for the detected neutrons.

MCNP model			Calculation results				
δm_{235}, %	Core radius × height, cm	H/U atom ratio	k_{eff}	Λ / Λ_0	$\varepsilon \times 10^4$	D_ν	λ_b / λ_{b0}
0 (design)	6.27 × 10.55	0.000	1.00004	1.000	8.108	0.8201	1.000
4	7.11 × 11.38	0.330	0.99980	1.779	7.330	0.8193	0.531
8	7.44 × 11.91	0.650	1.00055	2.479	6.588	0.8187	0.359
12	7.69 × 12.30	1.025	1.00095	3.229	5.853	0.8180	0.258

Another design alteration scenario considered modifications to the reflector. Modelling for the inner beryllium reflector, known to be extremely efficient in thermalizing fission neutrons and returning them back to the core, showed that the reactor could reach criticality even when nearly a third of the HEU was removed from the core (Table 2). The results indicated a significant (> 500-fold) increase in the prompt neutron generation time and a substantial drop (by a factor of ~10) in detection efficiency. This translates into a considerable drop of >1600 in the λ_b parameter compared to its design value, thereby confirming its very high sensitivity to design changes that entail neutron moderation.

Also shown in Table 2 are the results for another scenario that involves a partial replacement of the inner copper reflector with a low-grade fissile material. Due to the additional neutron multiplication inside such a fissile reflector, criticality can be maintained with a smaller amount of HEU in the reactor core. A series of simulations was performed to model 25%, 50%, and 100% replacements of the inner copper reflector with natural uranium (NU) metal.

Calculations for the actual core design showed that a complete replacement of the inner reflector with NU metal would create a significant reactivity margin (k_{eff} = 1.0149, ρ = +222 ¢). With this margin, criticality can be achieved even after the removal of 1.23 kg of ^{235}U (>5% of the nominal HEU core inventory). Across the range of simulated removal levels, the prompt neutron generation time would drop by ~30% compared to its design value, while the detector efficiency and Diven factor would stay nearly the same. As a result, the λ_b parameter would increase noticeably (by ~37%) compared to its nominal value.

Table 2. Results of the MCNP calculations for a modified TAPIRO reactor with Be and NU reflectors as would be needed to compensate for reactivity loss due to a hypothetical removal of various δm_{235} fraction of HEU from the core. The calculated Λ and λ_b values are shown as ratios to their values Λ_0 and λ_{b0} at the nominal reactor design. Detector efficiency was calculated assuming the low-level threshold of 44 keV for the detected neutrons.

MCNP model			Calculation results				
δm_{235}, %	Core radius × height, cm	Inner reflector	k_{eff}	Λ / Λ_0	ε ×10^4	D_ν	λ_b / λ_{b0}
30.3	6.27 × 5.72	Be	1.00107	530.1	0.797	0.8187	5.9·10^{-4}
1.15	6.27 × 10.43	NU (25%)	1.00010	0.901	7.977	0.8201	1.101
2.37	6.27 × 10.30	NU (50%)	1.00023	0.825	7.856	0.8200	1.193
5.30	6.27 × 9.99	NU (100%)	1.00022	0.710	7.630	0.8200	1.370

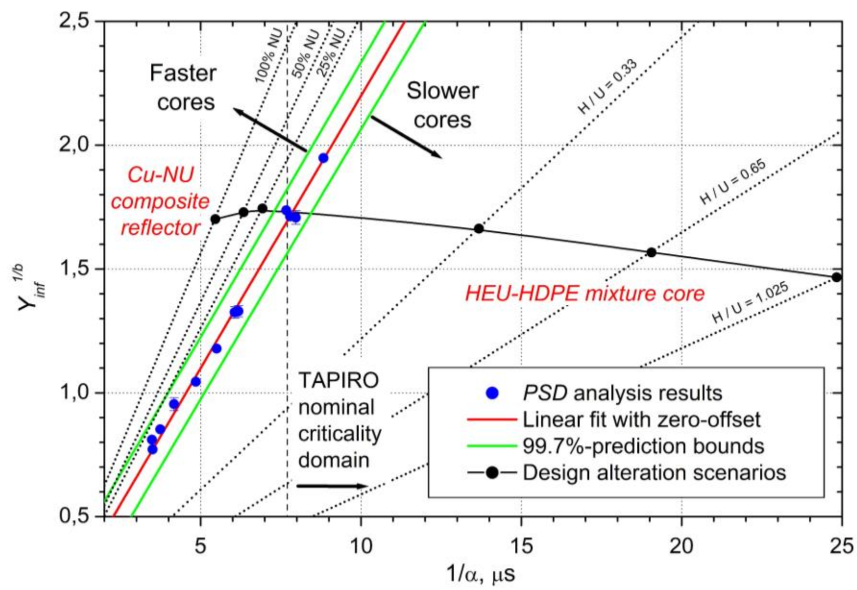

Figure 4. The $Y_\infty^{1/b}$ vs. α^{-1} plot combining the results of the PSD analysis of the neutron noise measurements with the results of the sensitivity study for two design alteration scenarios: (i) neutron moderation in a mixed HEU-HDPE core, and (ii) neutron multiplication in a composite Cu-NU reflector.

4.2. *Quantification of in-core nuclear material*

The results of the experimental measurements and the sensitivity study are combined in the $Y_\infty^{1/b}$ vs. α^{-1} two-dimensional plot shown in Figure 4. From the considerations in the previous section, a verification measurement performed at an arbitrary reactor criticality level can be expected to appear within certain acceptance limits, e.g., within the 3σ-prediction band as shown in Figure 4.

Obtaining a data point outside the acceptance limits would indicate a possible alteration of the reactor design to have led to a faster or slower core. The potential impact on safeguards objectives in this case can be estimated using the datasets in Table 1 and Table 2. These are plotted in Figure 5 in the form of a λ-calibration curve that defines the relationship between the measured m_{235} ^{235}U mass in the reactor core and the ratio of the measured and designed λ_b values:

$$\frac{m_{235}}{m_{235N}} = \begin{cases} 1 + 0.04582 \cdot \ln\left[1 - 1.262 \cdot (1 - \lambda_b/\lambda_{b0})\right], & \lambda_b/\lambda_{b0} < 1 \\ 1 + 0.11910 \cdot \ln\left[1 + 1.000 \cdot (1 - \lambda_b/\lambda_{b0})\right], & \lambda_b/\lambda_{b0} > 1 \end{cases} \quad \text{Eq. 5}$$

where, m_{235N} is the nominal ^{235}U mass. From Eq. 5, a measured λ_b value at one of the 3σ-prediction limits would indicate a potential decrease of the core inventory by $\delta m_{235} = 0.5\%$ ($\Delta m_{235} = 110$ g) or $\delta m_{235} = 0.9\%$ ($\Delta m_{235} = 200$ g^{235}U) in the case of design modifications leading to either a faster or slower core, respectively. It must be noted that these results were obtained for a critical reactor, where the sensitivity of λ_b to potential design alterations is expected to be maximal.

Once the reactor design has been verified based on the measured λ_b, the in-core ^{235}U mass can be evaluated using an α-calibration [3], which establishes a relationship between the ^{235}U mass and the α^{-1} value. The α-calibration curve and respective computational data for the TAPIRO reactor are shown in Figure 5 and Table 3.

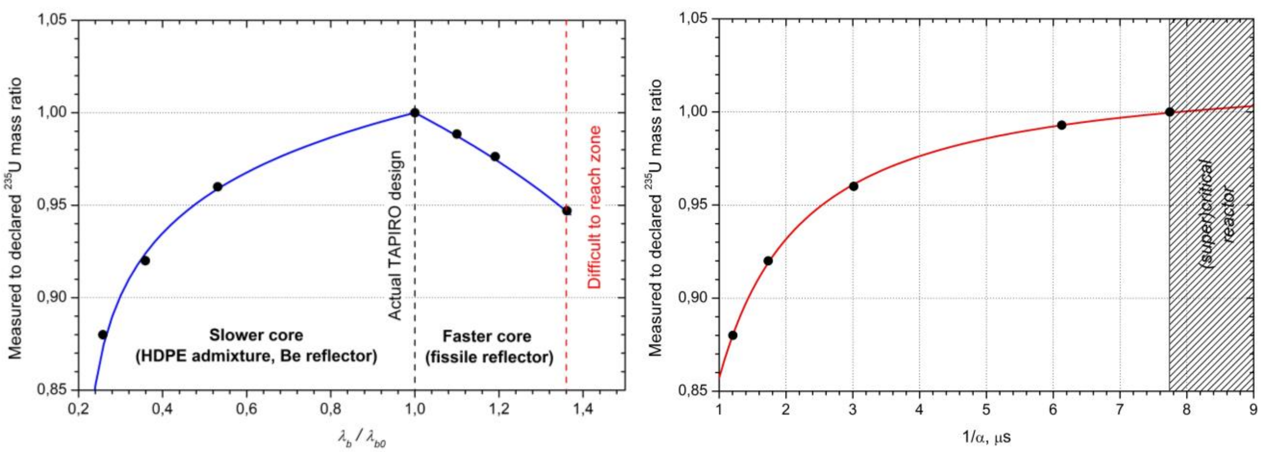

Figure 5. The λ-calibration (left) and α-calibration (right) curves for the TAPIRO reactor.

Table 3. Results of the MCNP calculations for different HEU loadings of the TAPIRO core. The calculated Λ and λ_b values are shown as ratios to their values Λ_0 and λ_{b0} at the nominal HEU loading. Detector efficiency was calculated assuming the low-level threshold of 44 keV for the detected neutrons.

MCNP model		Calculation results					
δm_{235}, %	Core radius × height, cm	k_{eff}	Λ / Λ_0	$\varepsilon \times 10^4$	D_ν	λ_b / λ_{b0}	α^{-1}, μs
0 (design)	6.27 × 10.55	1.00000	1.000	8.108	0.8201	1.000	7.747
4	6.27 × 10.13	0.98940	0.996	8.111	0.8201	1.004	3.016
8	6.27 × 9.32	0.97689	0.994	8.127	0.8201	1.007	1.736
12	6.27 × 8.20	0.96428	0.990	8.137	0.8201	1.012	1.206

The simulation results demonstrate a drastic change in the effective multiplication factor and α values when core loading is reduced. Across the simulated range of HEU loadings, the prompt neutron generation time, detector efficiency, Diven factor and, hence, λ_b, deviate only slightly from their nominal core values. In terms of the $Y_\infty^{1/b}$ vs. α^{-1} plot in Figure 4, it means that a neutron noise measurement result from a not fully loaded core would appear in the subcritical domain, within the 3σ-prediction band limits. The α-calibration curve is well described by the double exponential decay function:

$$\frac{m_{235}}{m_{235N}} = (1 - A_1\{1 - \exp(k_1[1 - \alpha_0\alpha^{-1}])\} - A_2\{1 - \exp(k_2[1 - \alpha_0\alpha^{-1}])\}) \qquad \text{Eq. 6}$$

where $A_1 = 9.84 \cdot 10^{-3}$, $A_2 = 2.41 \cdot 10^{-7}$, $k_1 = 2.61$, $k_2 = 14.26$, and α_0 refers to the nominal core loading. Using Eq. 6 and considering the 3σ-prediction interval $\Delta(\alpha^{-1}) = \pm 0.75$ μs (see Figure 4), the standard measurement uncertainty of the ^{235}U mass can be estimated to be: $\delta m_{235} = 0.08\%$ ($\Delta m_{235} = 18$ g^{235}U) and $\delta m_{235} = 0.14\%$ ($\Delta m_{235} = 32$ g^{235}U) for measurements in the critical ($k_{eff} = 1$) and closest-to-critical subcritical ($k_{eff} = 0.9982$) states, respectively.

The uncertainties appear to be extremely small. However, an unavoidable systematic bias due to the excess reactivity of the core must be considered, as the associated ^{235}U mass will create a negative offset of the measured vs. actual core loadings. In the case of the optimally designed, zero-power, fast-reflected core of TAPIRO, the offset is minimal: ~620 g^{235}U. Similarly, an additional offset must be considered for verification measurements performed in subcritical states. For the closest-to-critical subcritical state of TAPIRO (k_{eff} = 0.9982) examined herein, the subcritical-to-critical offset is about 158 g^{235}U, which is a small fraction of the total HEU core loading. Other potential sources of uncertainty due to instability in the detector and/or electronics or to reactor operational needs, e.g., removal of the reflecting plugs at the end of experimental channels for the purpose of sample irradiation, should be further investigated.

5. Conclusions

A partial defect test is proposed for the verification of difficult-to-access in-core nuclear material. The test is based on neutron noise measurements and data processing using the Feynman-α and PSD algorithms. Work done at the TAPIRO research reactor demonstrated that the derived neutron multiplication (Y_∞) and time evolution (α) characteristics could be utilized for design verification (λ-calibration) and in-core ^{235}U mass quantification (α-calibration).

The introduced reactor-specific λ_b parameter showed high sensitivity to reactor design changes, including modifications to the core and reflector, and independence of the criticality level, which makes it unique for design verification applications and for detection of concealment strategies (by their effects on reactor kinetics). The proposed verification methodology was able to determine the ^{235}U mass in the TAPIRO core with a random standard uncertainty $\delta m_{235} \leq 1\%$. Potential sources of systematic uncertainty include negative offset due to excess reactivity in the reactor core, measurement system instabilities, changes in experimental channels etc.

Amongst the benefits of the proposed methodology is its applicability to both critical and subcritical cores. Moreover, depending on the core status, verification can be performed in either subcritical, critical or supercritical states. Crucial for the successful implementation at a specific facility are the selection of a detector with a proper spectral response, timing characteristics and shielding and the possibility to install it in the vicinity of the core. Moreover, dedicated experimental and computational studies need to be done for selecting proper measurement locations, establishing reactor-specific parameters, fine-tuning data processing algorithms, determining acceptance limits and developing facility-specific measurement procedures.

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