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A Combined Computational and Measurement Approach for Safeguards Verifications of Subcritical Cores

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Abstract - *This paper describes a combined computational and measurement approach for quantitative determination of nuclear material mass in a subcritical system based on reactor noise analysis method. First application of the described methodology in support of international safeguards verifications is reported and its practical advantages and disadvantages are discussed.*

I. INTRODUCTION

The standard "criticality check" employed by IAEA inspectors to verify in-core nuclear material (NM) is based on the confirmation of the critical state of a reactor or a critical assembly. This is attained by observing the exponential increase in the neutron flux versus time upon insertion of a small positive reactivity into the system. In this way, the presence of at least one critical mass in the given configuration can be confirmed.

In recent years, there has been an increasing need to perform verifications of non-operational and shutdown reactors, for which the standard "criticality check" could not be applied. It was equally not applicable for the verification of subcritical cores, such as subcritical assemblies or accelerator driven systems, which have been becoming popular in the recent years. To address this need, an alternative approach for the verification of in-core nuclear material based on neutron noise analysis has been tested.

The neutron noise analysis is a well-known method [1] for reactivity measurement in subcritical systems based on the observed fluctuations in the neutron flux, representing a direct signature of fissile material. As such, it offers a wide applicability area and a great robustness with regard to possible diversions of NM. Furthermore, when combined with computational capability, the measured signal can be converted into the quantity of nuclear material, provided that the system design is known and verifiable, and its state during verification measurements is properly controlled.

The paper presents details on the practical implementation of this combined computational and measurement approach and reports on its application to the confirmation of the presence and verification of the quantity of highly-enriched uranium in the shutdown IIN-3M reactor in Tashkent, Republic of Uzbekistan.

II. DESCRIPTION OF THE ACTUAL WORK

1. Theoretical

Reactor noise analysis has been widely applied for reactivity measurement of sub-critical systems [1]. In the current paper, a particular implementation of the method represented by the Feynman variance-to-mean approach [2] (also known as Feynman-α analysis), is discussed. In this approach, fluctuations in the number of neutron counts detected within a time interval of length t are expressed as the ratio of the variance of number of counts $\sigma_Z^2(t)$ to the mean number of counts $\langle Z(t)\rangle$. This ratio is further referred to as the variance-to-mean ratio (VMR). At time intervals $t \leq 1$ s, the excess of the VMR with respect to unity can be expressed as a simple function of the detector and reactor core parameters [1, 2]:

$$Y(t) = \frac{\sigma_z^2(t)}{\langle Z(t)\rangle} - 1 = \varepsilon A \times \left(1 - \frac{1-\exp(-\alpha t)}{\alpha t}\right) \quad (1)$$

$$A = \frac{D_\nu}{(\beta-\rho)^2}, \quad \alpha \approx \frac{\beta-\rho}{\Lambda}, \quad \rho \approx \frac{k_{eff}-1}{k_{eff}} \quad (2)$$

Here, ε is the detector efficiency, β is the effective delayed neutron fraction, Λ is the prompt neutron generation time, ρ is the reactivity, D_ν is the Diven factor that depends on the fission neutron multiplicity ν, and k_{eff} is the effective neutron multiplication factor.

Several approaches to inferring NM mass in a sub-critical core based on Eqs (1) and (2) can be considered. For instance, based on experimentally obtained function $Y(t)$, parameter α can be determined. The, provided that the core specific parameters β and Λ are known, values of ρ and k_{eff} can be estimated. These can be compared with predicted values, which can be computed as function of the NM mass, e.g. using a Monte Carlo method. Thereby the amount of NM in the core can be evaluated.

A seems to be more robust approach would rely on the comparison of measured and predicted values of parameter α, which is a strong function of the NM mass. This approach is more favorable, as it eliminates potential influences from the (unknown) core specific parameters β and Λ, which are now included in the integral parameter α.

Another approach would be to compare experimentally obtained and theoretically calculated values of $Y(t)$ at certain time interval length, e.g. at very long time intervals $t\to\infty$. At such times, function $Y(t\to\infty) = Y_\infty$ does not depend on the neutron prompt generation time, however it carries additional dependence on fissile material (described by the first and second moments of the multiplicity distribution of fission neutrons present in the definition of the Diven factor) and also depends on the detector efficiency.

Examples of application of the above approaches will be given in the following sections. Generally speaking, all these represent one way of circumventing a complex inverse problem by means of testing the consistency between different measurable observables and respective predicted values. Appropriateness of this approach in case of safeguards verifications has been discussed recently in [3].

2. Computational

To support the described verification approaches, Monte Carlo computations were carried out with the help of MCNPX 2.6.0 code [4] and by means of a modified version of its predecessor code - MCNP4c. The ENDF/B-VII.0 library was the main source of the neutron cross-section data, complemented by the ENDF/B-VI.6 when necessary. In case of the modified MCNP4c, the LLNL's fission library [5] was adopted for accurate simulation of spontaneous and induced fission events. This simulation option was utilized for the results presented in Section III.

Criticality calculation is one of the standard features of the MCNP codes, yielding values of k_{eff} that could be directly compared with measurement results. Both MCNP codes were also capable of generating detector pulse trains, e.g. using the PTRAC feature or via direct logging of progressively increasing time intervals associated with neutron detection events. The obtained pulse trains normally containing up to 10^6 events were subsequently analyzed using custom made post-processing software, which built and extracted parameters of the modeled $Y(t)$ distributions and could also simulate the counter dead time effects.

Creation and validation of an MCNP model for a verified subcritical core constituted one of the most crucial tasks that contributed significantly to the final accuracy of verification. Models could be created based on design drawings, which were normally available from a facility operator. These were verified and, if needed, complemented by on-site measurements and visual observations.

The created models were benchmarked by comparing calculated values of k_{eff} with nominal or experimentally determined values stated in reactor or assembly passport. The comparisons were performed at different reactivity levels, e.g., at different positions of the control and/or safety rods.

3. Experimental

Measurements were performed using un-moderated ^{3}He and ^{10}B lined proportional neutron counters from GE Reuter Stokes (USA), which were coupled to a charge sensitive preamplifier from Precision Data Technologies Inc. (USA). Two different setups were utilized for data acquisition.

One of them employed InSpector 2000 DSP Portable Spectroscopy Workstation from Canberra Industries (USA). This was used to record multichannel scaling (MCS) spectra with ≥ 5 ms fixed dwell time (i.e., length of the time interval t) and <10 μs dwell time resolution. The obtained MCS spectra were processed to extract VMR values and their uncertainties.

Another setup was based on the PTR-32 multichannel pulse train recorder developed in the Institute of Isotopes (KFKI, Hungary). This device allowed recording sequences of detector pulses at up to 2.5 Mcps input count rate and with time stamping resolution ±5 ns. The recorded data arrays were analyzed by a dedicated post-processing code, to infer parameters of the Feynman-α distributions.

Detectors were positioned in close proximity to or inside a core, e.g. in one of experimental channels. In case of start-up source, detector position was chosen such that to minimize direct exposure by the source neutrons. Series of measurements were performed with and without start-up source as well as at different positions of safety and/or control rods.

Visual observations and additional measurements, e.g. using a collimated gamma-ray detector to confirm location and dimensions of the core via measured radiation profile of the core, were essential parts of field activities that corroborated correctness of the provided design information and confirmed declared status of the verified subcritical system.

III. RESULTS

This section presents results of application of the described methodology for quantitative measurement of NM amounts in support of the IAEA's safeguards verification activities during unloading of the fuel from the IIN-3M research reactor.

The IIN-3M reactor was located in Tashkent, Republic of Uzbekistan, and operated by the Joint Stock Company "Photon" from 1975 until 2013. In 2013 the reactor was shut down and its fuel was destined to shipment to Russian Federation within the Russian Research Reactor Fuel Return Programme sponsored by the U.S. NNSA.

The IIN-3M represented a type of self-extinguishing pulse reactors capable of producing large power bursts resulting from an abrupt change in the reactor reactivity from sub-critical to prompt critical [6]. The 40 cm diameter high-pressure vessel contained nominally 22.8 liters of uranyl sulfate water solution with 90 wt% ^{235}U enrichment, 200 gU/l concentration and 1.28 g/cm^3 density. The reactor was regulated using four control rods and one safety rod, all made of boron carbide (B_4C).

Schematic MCNP model of the reactor, showing also the 2 atm ^{3}He neutron counter positioned inside reactor's central experimental channel, is shown in Fig. 1. The model was created based on design information and validated by comparing the calculated multiplication factors against available passport data: k_{eff} (calc) = 1.0353(16) vs k_{eff} (pass) = 1.035 with all control rods withdrawn; k_{eff} (calc) =

0.9646(15) vs k_{eff} (pass) = 0.963 with all control rods inserted.

Verification of the reactor design was performed by means of a visual inspection and also experimentally by measuring vertical profiles of the reactor core using a highly collimated 60 mm^3 CdZnTe gamma-ray detector (Fig. 2).

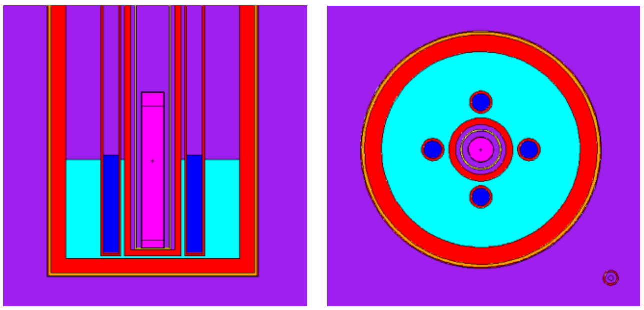

Fig. 1. MCNP model of the IIN-3M reactor: cyan – uranyl sulphate water solution; blue – B_4C; red – stainless steel; yellow – water; magenta – ^{3}He gas.

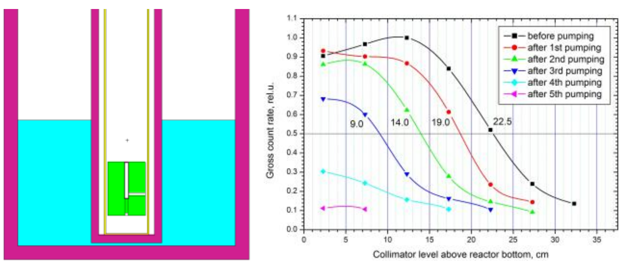

Fig. 2. Measurement geometry (on the left) and results of the gamma-ray profiling of the IIN-3M reactor core at different stages of reactor fuel discharge (on the right). The numbers shown indicate fuel fill heights determined.

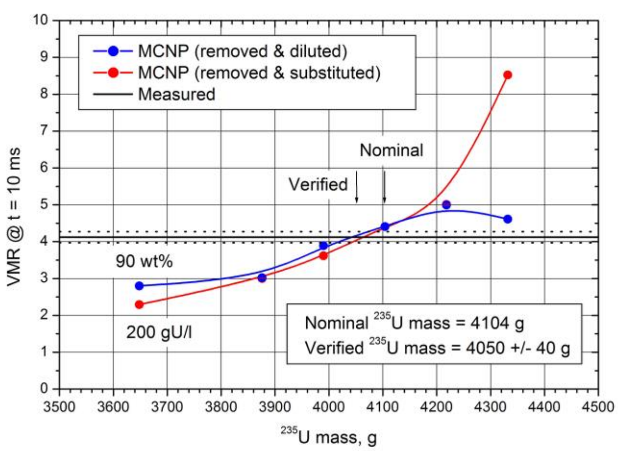

Fig. 3. Measured vs. calculated VMRs for a fully loaded IIN-3M reactor. Blue and red dots show results of MCNP calculations for diversion scenarios (a) and (b), respectively. Computational precision is within the size of the dots.

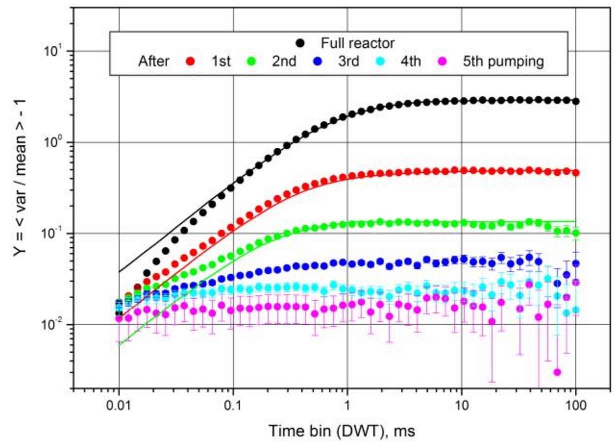

Fig. 4. Feynman-α distributions built using measured pulse trains. Solid lines connect MCNP calculation data points for the full reactor (black line), after 1st pumping (red line), and after 2nd pumping (green line). Computational precision is negligible compared to the size of experimental data points.

Comparison of the measured vs. simulated profiles was essential for the correct interpretation of the data that yielded 22.5 ± 0.5 cm and 22.8 ± 0.5 l for the fuel level and volume, respectively. The experimentally determined fuel level was adopted in the final MCNP model of the IIN-3M reactor, which was essential for the correct interpretation of neutron data.

The VMR values were calculated as function of the ^{235}U content assuming following diversion scenarios:

(a) Partial removal of NM with subsequent dilution by water that would result in less concentrated fuel with preserved original enrichment level, and

(b) Partial removal of NM with subsequent substitution using lower enriched material that would result in lower enriched fuel with the original uranium concentration.

Fig. 3 compares results of the predictive calculations for both diversion scenarios with experimentally determined VMR at t = 10 ms. The comparison yielded 4050 ± 40 g for the ^{235}U mass and its 1σ-uncertainty, which was in reasonable agreement with the nominal value and the NM amount declared by the facility operator.

Further measurements were performed during unloading of the fuel, which was performed in five cycles. In order to control the amount of material transferred during the cycles, gamma profiling and neutron measurements were performed after completion of each cycle. All neutron data were acquired for 1000 sec measurement time.

The measured gamma-profiles shown in Fig. 2 suggested a stepwise decrease of the fuel level in the course of first three cycles by Δh = 4.5 ± 0.9 cm, which corresponded to $\Delta V = 4.6 \pm 0.9$ l or $\Delta m = 830 \pm 160$ g^{235}U per cycle and was in agreement with level meter readings that were available from reactor control panel. Measurements beyond the 3th cycle were not conclusive as the fuel could not be seen anymore by the detector.

The measured vs. simulated Feynman-α distributions are shown in Fig.4. These agree very well at t > 0.5 ms, however show noticeable deviations at shorter time intervals, thereby pointing to possible influences from unaccounted instrumental effects and inconsistencies between actual and modeled reactor layouts. It is also noteworthy that after the 3th cycle, when more than half of the fuel had been removed, Feynman-α distributions started suffering considerably from the lack of count statistics.

The measured and calculated Y(t = 10 ms) and 1/α at different fuel levels are shown in Fig. 5 and Fig. 6, respectively. Intersections of the horizontal lines, representing measured values, with theoretical curves gave estimates of the fuel levels and hence of the ^{235}U masses. These were consistent with the results of gamma-profiling shown in Fig. 2. Again, application of this approach was inefficient after 3rd cycle due to substantial increase of the measurement uncertainty.

IV. CONCLUSIONS

In this work, for the first time, the reactor noise analysis was employed for the determination of NM mass in a subcritical system in support of safeguards verifications. Amongst several approaches tested, the use of the measurable parameter α for inferring unknown NM mass looks more adequate, as application of this approach does not require knowledge of additional reactor core specific parameters, such as prompt neutron generation time and effective delayed neutron fraction. It is also free from the influences of detector efficiency and neutron source strength, which normally require special considerations in case of neutron coincidence counting (NCC).

Practical utilization of the approach requires however substantial effort towards creation and validation of a computational model as well as for reactor/assembly design information verification. The approach also becomes inefficient in case of very deep subcritical systems exhibiting very low neutron multiplication. Here safeguards traditional active NCC methods stay more appropriate.

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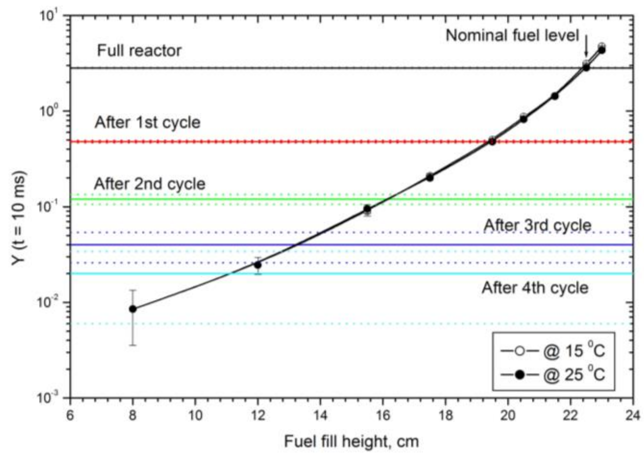

Fig. 5. Y(10 ms) as function of fuel level: solid curves - MCNP calculation results; solid and dotted horizontal lines - experimental values and their 1σ confidence intervals at different stages of reactor de-fuelling.

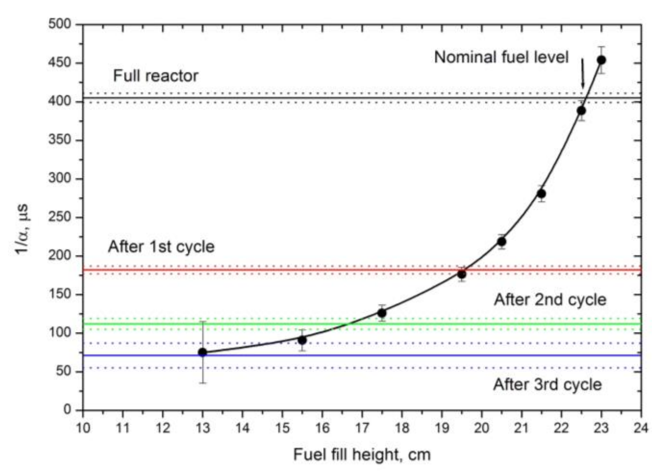

Fig. 6. Parameter 1/α as function of the fuel level. Solid curve shows results of MCNP calculations. Horizontal lines represent experimental data - mean values (solid lines) and 1σ confidence bands (dotted lines) at different stages of reactor de-fuelling.