CHARACTERISATION OF NUCLEAR POWER PLANT SPENT FUELS FOR SAFEGUARDS USING IN-SITU GAMMA SPECTROMETRY

PhD thesis booklet

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Background of the research

The Nuclear Security Department (NSD) of the HUN-REN Centre for Energy Research has been studying the spent fuel assemblies of Paks Nuclear Power Plant (NPP) Ltd. in Hungary over the last two decades. Following the 2003 serious incident, the NPP requested the quantification of nuclear material in canisters containing spent fuel debris. Gamma- and neutron-based measurements were employed, and using intact spent fuel assemblies as reference, researchers from the NSD successfully determined the nuclear material content of each canister [Zsigrai, 2013]. Leveraging their experiences with the canister measurements from 2010, the researchers continued with the gamma-ray spectrometry measurement of spent fuel assemblies, carrying out yearly data acquisition campaigns. The goal of these studies is to validate the burnup level of spent fuels for the assessment of the true operator-calculated burnup uncertainty [Nguyen, 2013]. The collected experimental dataset has already served as a basis for some previous research, which was aimed at the analysis of spent fuel operational history [Kocsonya, 2016] [Kirchknopf, 2018] [Kirchknopf, 2019].

The topic of my PhD dissertation is the determination of spent fuel parameters relevant to nuclear safeguards from gamma-ray spectrometry measurements carried out on VVER-440 reactor type assemblies. The parameters are the burnup, cooling time, initial enrichment, remaining fissile material content, and the activities of detectable fission products. There exist many scientific papers that study a similar problem for different kinds of spent fuels, but few of them consider specifically the VVER-440 type, which was the focus of my work. Researchers have often used regression analysis to calculate the relevant spent fuel parameters, but aside from this technique, one of my main goals was the implementation of various machine learning methods to achieve more reliable calculations [Favalli, 2016] [Hellesen, 2017] [Rossa, 2020].

References:

J. Zsigrai et al., „Non-Destructive Determination of the Nuclear Material Content of Spent Fuel Pieces in Canisters”, *IEEE Transactions on Nuclear Science*, 60(2), pp. 1080-1085, 2013.

C. T. Nguyen et al., „Monitoring Burn-Up of Spent Fuel Assemblies by Gamma Spectrometry”, *IEEE Transactions on Nuclear Science*, 60(2), pp. 1107-1110, 2013.

A. Kocsonya, „Kiégett fűtőelemek előéletének meghatározása gamma-spektrometriával (1. év)”, OAH-ABA-19/16-M Research report (MTA EK SBL), 2016.

P. Kirchknopf & A. Kocsonya, „Kiégett fűtőelemek előéletének meghatározása gamma-spektrometriával (2. év)”, OAH-ABA-20/17-M Research report (MTA EK SBL), 2018.

P. Kirchknopf, I. Almási, Z. Hlavathy & P. Völgyesi, „Kiégett fűtőelemek előéletének meghatározása gamma-spektrometriával (3. év)”, OAH-ABA-24/19-M Research report (MTA EK SBL), 2019.

A. Favalli et al., „Determining initial enrichment, burnup, and cooling time of pressurized-water-reactor spent fuel assemblies by analyzing passive gamma spectra measured at the Clab interim-fuel storage facility in Sweden”, *Nuclear Instruments and Methods in Physics Research A*, 820, pp. 102-111, 2016.

C. Hellesen et al., „Nuclear spent fuel parameter determination using multivariate analysis of fission product gamma spectra”, *Annals of Nuclear Energy*, 110, pp. 886-895, 2017.

R. Rossa et al., „Comparison of machine learning models for the detection of partial defects in spent nuclear fuel”, *Annals of Nuclear Energy*, 147, p. 107680, 2020.

Objectives

The most important objective of my research is to develop novel methods for safeguards relevant parameter calculations (burnup, cooling time, initial enrichment, and fissile material content) of VVER-440 type spent fuel assemblies. The existence of a rapid and easy-to-use, as well as accurate method of spent fuel parameter measurement would improve the abilities of safeguards inspectors to detect potential violations of safeguards agreements and violations of the principle of nuclear non-proliferation. The relevance of this work is underlined by the incomplete arsenal of the currently used safeguards verification methods for spent fuels, because in most cases today, only the mere presence of fissile material and not the quantity or isotopic composition is checked, usually by a Cherenkov viewing device.

Developing a spent fuel parameter measurement method is also relevant from a nuclear forensic perspective. During the nuclear forensic investigation of a spent fuel sample, the analysts are tasked with the measurement of all key signatures that are characteristic of that sample and are related to its origin. All of this is to be carried out without any prior knowledge of the sample, i.e., no cooling time or other information is known beforehand. Finding a solution to this problem yields a measuring and calculation method that would allow the nuclear forensic analysts to characterise pieces of spent fuel of unknown origin, leading to increased capabilities in the field of nuclear security.

Finally, it is an important objective to evaluate the burnup calculation uncertainty of the NPP, which would, in theory, allow the reduction of the burnup engineering factor that limits the total power output of an assembly. Achieving this goal would result in more economical fuel usage, while nuclear safety remains uncompromised.

Information about the fission product content is also relevant to the NPP operators and to national authorities from a nuclear security and radiation protection perspective. The development of a reliable measurement and evaluation technique makes it possible to determine certain fission product activities. Carrying out dose calculations would be possible from the measurement of fission product activities. Moreover, radiation-induced damage in the structure surrounding the spent fuel assemblies could be studied based on these measurements. The relevance of this objective is supported by the fact that at the NPP, such experimental studies were not carried out before.

Methods

I used gamma-ray spectrometry as the experimental technique to non-destructively assay spent fuel assemblies. Measurements of the extremely active fuel assemblies were done in thick water shielding and through a long and narrow collimator. The resulting gamma energy spectrum in the range of 50-3000 keV is from a vertically thin slice of the assembly and contains only fission product gamma peaks. A limitation of this method is that the gamma-rays of fissile isotopes cannot be detected due to their low energy and intensity compared to the fission products.

Following the spectrum evaluation, I calculated the activity ratios of the fission product isotopes that were detected in the spent fuel assemblies. I employed non-linear regression between the measured activity ratios and the spent fuel parameters to study their observable correlations. To obtain further information embedded in the measured spectra, I applied various machine learning algorithms with the intent to improve the calculation accuracy and to determine the operational history of the fuels. The calculation of fission product activities was done with an efficiency calibration based on Monte Carlo particle transport, for which I constructed the simulation model of the whole experimental setup.

New scientific results

[T1] Thesis: Burnup prediction using fission product activity ratios measured with gamma spectrometry

I determined empirical relations between fission product activity ratios measured by gamma spectrometry and the burnup of VVER-440 type spent fuels. By regression analysis with a power function model, I concluded that for spent fuels with less than 9 years of cooling time, the $^{134}Cs^2/(^{106}Ru^{137}Cs)$ activity ratio yields, on average, nearly five times more accurate burnup prediction than the widely used $^{134}Cs/^{137}Cs$ activity ratio. I showed that it is possible to use the $^{134}Cs^2/(^{106}Ru^{137}Cs)$ activity ratio for burnup prediction even without the knowledge of the cooling time, with only a 19% reduction in prediction accuracy [1].

[T2] Thesis: Cooling time prediction and the determination of origin with gamma spectrometry

I showed using exponential model functions that the cooling time of VVER-440 type spent fuel assemblies cooled between 0.5-5 years can be predicted using the measured $^{110m}Ag/^{134}Cs$ activity ratio with an average ±3 months uncertainty. Using the same measurement and calculation methods for cooling times in the range of 0.5-12 years, the application of the $^{134}Cs/^{154}Eu$ activity ratio yields an accuracy of ±4 months. I concluded that for cooling times of less than 6 months, the activities of the ^{136}Cs, ^{140}Ba, and ^{156}Eu isotopes relative to the activity of ^{137}Cs function as indicators for the shortness of the cooling time. I pointed out that the reliable determination of temporal origin by the $^{134}Cs/^{154}Eu$ and $^{110m}Ag/^{134}Cs$ activity ratios is only possible if the reactor unit of origin is known [1].

[T3] Thesis: Parameter prediction and operational history investigation with machine learning approaches

I conducted studies with the application of machine learning models to increase the parameter prediction accuracy. The calculation error of the burnup saw a marginal decrease, while for the cooling time, it fell back nearly to a third with the best performing models. As opposed to the regression technique, the prediction of the initial enrichment proved successful, and I showed that the ^{239}Pu concentration can be predicted with good accuracy using neural networks. A novel achievement is the parameter prediction carried out directly from the gamma spectrum using a convolutional neural network. Using cluster analysis, I concluded that the variation of the per-cycle burnup operational history is reflected in the activity ratios measured by gamma spectrometry [1] [2].

[T4] Thesis: Activity measurement using Monte Carlo simulation

I developed a novel method to calculate the activities of gamma-emitting fission products in spent fuel assemblies. Using Monte Carlo particle transport simulations, I accomplished the absolute detection efficiency calibration of the spent fuel gamma spectrometry measurements. I applied X-ray radiography and determined the germanium dead layer thickness with precise in-laboratory measurements to refine the simulation model constructed for the MCNP6.2 code. I validated the model in an indirect manner, using experimentally calculated relative detection efficiencies and VVER-440 spent fuel ^{137}Cs concentration data available in the SFCOMPO database. The ^{137}Cs activity calculated from this model is expected to agree with the true value within a –10% to +20% interval [3].

Scientific publications related to the thesis statements

[1] **Péter Kirchknopf**, István Almási, Gábor Radócz, Imre Nemes, Péter Völgyesi, Imre Szalóki, „Determining burnup, cooling time and operational history of VVER-440 spent fuel assemblies based on in-situ gamma spectrometry at Paks Nuclear Power Plant", *Annals of Nuclear Energy*, 170, p. 108975, 2022, https://doi.org/10.1016/j.anucene.2022.108975.

[2] **Péter Kirchknopf**, Bálint Batki, Péter Völgyesi, Zoltán Kató, Imre Szalóki, „Application of machine learning methods for spent fuel characterization based on gamma spectrometry measurements", *Annals of Nuclear Energy*, 205, p. 110601, 2024, https://doi.org/10.1016/j.anucene.2024.110601.

[3] **Péter Kirchknopf**, Zoltán Kató, Csongor Kristóf Szarvas, Péter Völgyesi, Imre Szalóki, „Monte Carlo based absolute efficiency calibration of power reactor spent fuel NDA measurements", *Annals of Nuclear Energy*, 211, p. 110953, 2025, https://doi.org/10.1016/j.anucene.2024.110953.

Other scientific publications

[4] Szarvas Csongor Kristóf, **Kirchknopf Péter**, Almási István, Dósa Gergely, Szabó Katalin Zsuzsanna, Nguyen Cong Tam, Völgyesi Péter, Radócz Gábor, Szalóki Imre, „Alacsony hátterű gamma-spektrometriai mérőrendszer HPGe detektorának modellezése törvényszéki és in-situ analitikai célokra", *Nukleon* XIV, p. 238, 2021, https://nuklearis.hu/sites/default/files/nukleon/14_1_238_Szarvas_0.pdf.

[5] M. Travar, J. Nikolov, N. Todorović, A. Vraničar, P. Völgyesi, **P. Kirchknopf**, I. Čeliković, T. Milanović, D. Joković, „Detailed optimization procedure of an HPGe detector using Geant4 toolkit", *Journal of Radioanalytical and Nuclear Chemistry*, 332, pp. 817-828, 2023, https://doi.org/10.1007/s10967-023-08810-x.

[6] Csongor Kristóf Szarvas, Radócz, Gábor, Anita Gerényi, György Szabados, Tamás Pintér, Péter Rozmanitz, Péter Kirchknopf, Imre Szalóki, „Time-dependent leakage model for the identification of defective fuel assemblies of VVER-type nuclear reactors. Part 2: Expediting the solution and applications to a nuclear power plant", *Annals of Nuclear Energy*, 197, p. 110234, 2024, https://doi.org/10.1016/j.anucene.2023.110234.