DEVELOPMENT OF A REACTOR MONITORING PROGRAMME FOR IN-LINE REACTOR FUEL RELIABILITY ASSESSMENT

I.A. MEULENBELT, L.G. DAVIS
High Flux Reactor (HFR), Nuclear Research & Consultancy Group
Westerduinweg 3, 1755 LE Petten – NETHERLANDS

ABSTRACT

This paper details the development of a Reactor Monitoring Programme using periodic gamma-ray spectrometry measurements of the primary coolant water. It is designed to monitor the performance and condition of reactor fuel and components, detect fuel-related anomalies such as fuel defects, and provide insights into the ageing of primary system components. The programme was originally designed for the High Flux Reactor in Petten, but the methodology is adaptable to other reactors, most easily for those using U_3Si_2-Al fuel. The programme is based on three components: establishing an Isotopic Baseline based on historical gamma-ray spectrometry data of HFR's primary coolant water, a Python-based Release Model for predicting fission product release from the fuel matrix, and the identification of Fuel Defect Indicators based on the Release Model. The resulting programme can assist analysts in interpreting the weekly gamma-ray spectrometry data allowing for enhanced reactor performance assessment.

1. Introduction

The High Flux Reactor (HFR) in Petten employs weekly in-line gamma-ray spectrometry (GRS) analyses of the primary coolant water (PCW) to determine its radiochemical composition. Previously, the interpretation of the weekly GRS data relied on expert judgement without quantitative backing. Although this practice has been adequate in the past, it lacks support for the evaluation of long-term trends. To address this limitation, the development of a Reactor Monitoring Programme (RMP) is introduced, initially designed for the HFR but adaptable to other Light Water Reactors (LWRs) due to the flexibility of its underlying components. The RMP supports expert assessment of weekly GRS measurements, enabling detailed monitoring of reactor fuel condition and performance, and includes capabilities for long-term trending and integrity assessment of primary circuit materials which may provide insights for ageing management programmes. It is designed for ease of use, only requiring users to input the weekly GRS measurement data in the frontend to instantaneously generate an analysis of the isotopic activity profile for the given period.

The RMP's backend consists of three key components: the Isotopic Baseline, the Release Model, and the Fuel Defect Indicators. The Isotopic Baseline, derived from historical data of normal steady-state operation, serves as a reference for comparing new weekly GRS results, enabling the identification of anomalies. The Release Model, a Python-based tool, models the diffusion of fission products from the fuel matrix through the cladding and the subsequent release into the PCW. This can aid in defining what radioisotope activity could be related to a fuel defect and not by typical isotope diffusion. This leads directly to the definition of fuel specific Fuel Defect Indicators, thus providing a quantitative basis for identifying potential fuel defects. Defining these indicators facilitates early detection of potential fuel defects.

2. Theory

The HFR in Petten is a 45 MW tank-in-pool light water cooled and moderated nuclear research reactor located in the Netherlands which achieved first criticality in 1961. The reactor contributes significantly to radioisotope production, primarily for medical application, as well as research and qualification of nuclear materials and fuels [1]. The low enriched uranium (LEU) fuel that is used in the HFR is ~20% enriched uranium silicide dispersed in aluminium (U_3Si_2-Al), with an aluminium alloy (AG3NE) cladding [2]. The HFR in core structural components and primary circuit are made primarily from aluminium alloys (5xxx and 6xxx) and to a lesser extent stainless steel [3-5]. Cadmium is employed in the control rods as neutron absorber. A global visualisation of the HFR layout is given in Figure 1.

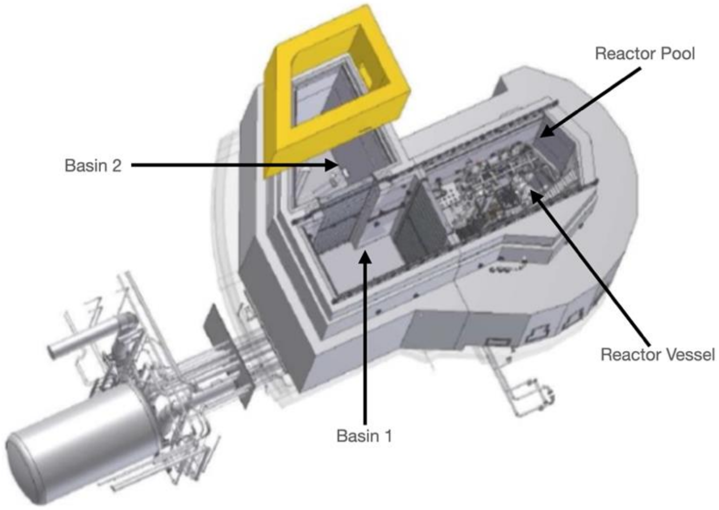

Figure 1: Schematic layout of the HFR Petten. The position of the storge and handling basins 1 and 2, reactor vessel, and reactor pool are indicated by arrows.

2.1. Release of Isotopes in the Primary Coolant Water

The GRS measurements detect and quantify radioisotopes present in the PCW [6]. There are several mechanisms by which isotopes can enter the bulk primary coolant water:

- Structural material-related elements may enter via corrosion and erosion processes of primary circuit components and/or reactor utilisation related system (isotope production and experimental facilities) components.
- Contaminant elements may be introduced into the primary circuit during reactor stop periods. Potential contaminants include tramp uranium from new fuel plates, chemical contaminants from tools, equipment, and lubricants used during maintenance and operational activities during refuelling reactor stops. Additionally, contaminants such as antimony may leach from polymer materials used in isotope production facilities.
- Volatile fission products, such as noble gases (e.g., Kr, Xe) and other volatile isotopes (e.g., I) [7, 8], may diffuse through the fuel cladding and/or its natural design porosities [9, 10].
- Release from the fuel matrix into the PCW due to a fuel defect [11]. A fuel defect occurs when the cladding of the fuel is degraded or penetrated, enabling a wide range of fission products to be released from the fuel matrix.

These release mechanisms lead to an array of isotopes potentially present in the PCW. These may be inherently unstable or may undergo neutron activation to a gamma emitting unstable radioisotope, making their activation products detectable by means of GRS. Information pertaining to the fuel and integrity of structural components may thus be elucidated by the measured radioisotope activity profiles in the PCW.

Certain isotopes are significantly affected by the pH of the PCW, and thus only detected in extended reactor stop periods during which the pH of the PCW is naturally reduced. This pH reduction during reactor stop, is as a result of the PCW mixing with the basin water when the reactor vessel (part of the primary circuit) is opened. The basin water is in constant contact with air in the reactor containment building, leading to the dissolution of atmospheric carbon dioxide into basin water (and the PCW during reactor stop), forming carbonic acid [12]. Carbonic acid, being a weak acid, decreases the pH and acts as an oxidiser, increasing the redox potential (Eh). Stainless steel components in the primary circuit feature a passivating layer of chromium oxide (Cr_2O_3). At nominal operating conditions with $pH > 6.5$, Cr_2O_3 is primarily solid state (mostly insoluble), thus the passivating film is solid and well adhered to the steel structural surface. When the Eh increases and the pH decreases, as in reactor stop, the solubility of Cr_2O_3 increases, releasing the Cr-51 accumulated in this film [13].

3. Methodology

The current procedures for GRS of the PCW have been in place since the start of 2022, following process improvements, optimisations, and equipment updates. Therefore, the GRS data from 2022 and 2023 was used for the initial development of the RMP.

3.1. Defining the Isotopic Baseline

The Isotopic Baseline quantifies the radioisotope activity profile in the PCW during normal steady-state operation. Before the quantitative baseline was established, the detected radioisotopes were categorised into isotopic groups displaying similar characteristics and/or of similar origin. The groups were defined as 'Structural Components', 'Contaminants', 'pH/Eh Related Products', and 'Volatile Fission Products'.

Periodic variation in group specific isotopic activity across the 28-30 days (4 weeks) operational period of the reactor necessitated defining a weekly baseline to improve baseline accuracy and reduce the confidence interval, as illustrated in Figure 2. The baseline is subsequently calculated for each week, for each radioisotope group using historical data, indicative of normal steady-state operation levels for each radioisotope group. This involves normalising individual radioisotope activity via min-max normalisation, followed by group-level normalisation based on the number of radioisotopes within each group.

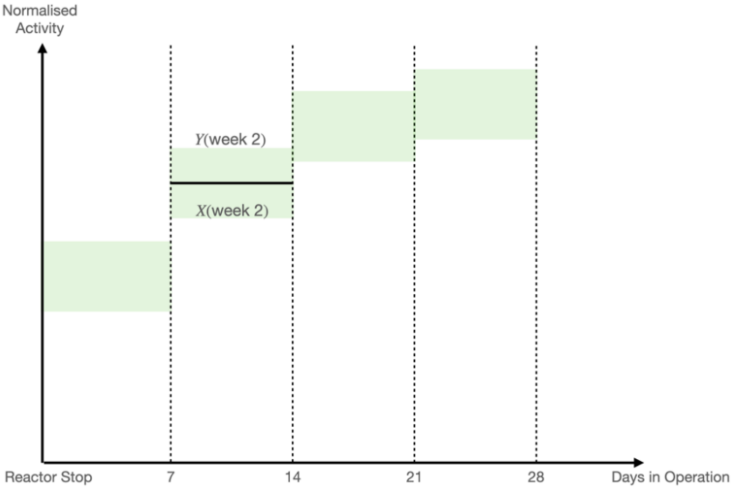

Figure 2: Visualisation of the baseline margins for a weekly defined baseline.

Based on this structure, the isotopic baseline can be formulated as Equation (1).

$$X(week) \le \sum_{i=1}^{n} \frac{A_i}{n} \le Y(week) \tag{1}$$

Where *n* represents the number of radioisotopes in a specific group, A_i the normalised activity of an isotope, and *X(week)* and *Y(week)* the respective lower and upper limits of the isotope group activity based on available data. Should a weekly GRS measurement fall outside of the confidence interval range defined by the Isotopic Baseline, with the normalised activity of a group falling either below X(week) or exceeding Y(week), the RMP automatically flags this anomaly, identifying the affected isotope group and whether its activity is anomalously high or low.

3.2. Development of the Release Model

To better understand the origin of the radioisotopes in the 'Volatile Fission Products' group and to support the identification of Fuel Defect Indicators, a Release Model was developed using Python. This model employs a modified version of Fick's second law, which integrates the decay of fission products, for accurate fission product transport modelling [14]. The Release Model was used to derive a Release Coefficient for Xe from the available GRS data. This coefficient replaces the diffusion coefficient in Fick’s second law as it is similar to the diffusion coefficient, but instead of only accounting for diffusion, the Release Coefficient encompasses all relevant mechanisms of isotope release, including diffusion enhanced by irradiation and recoil effects.

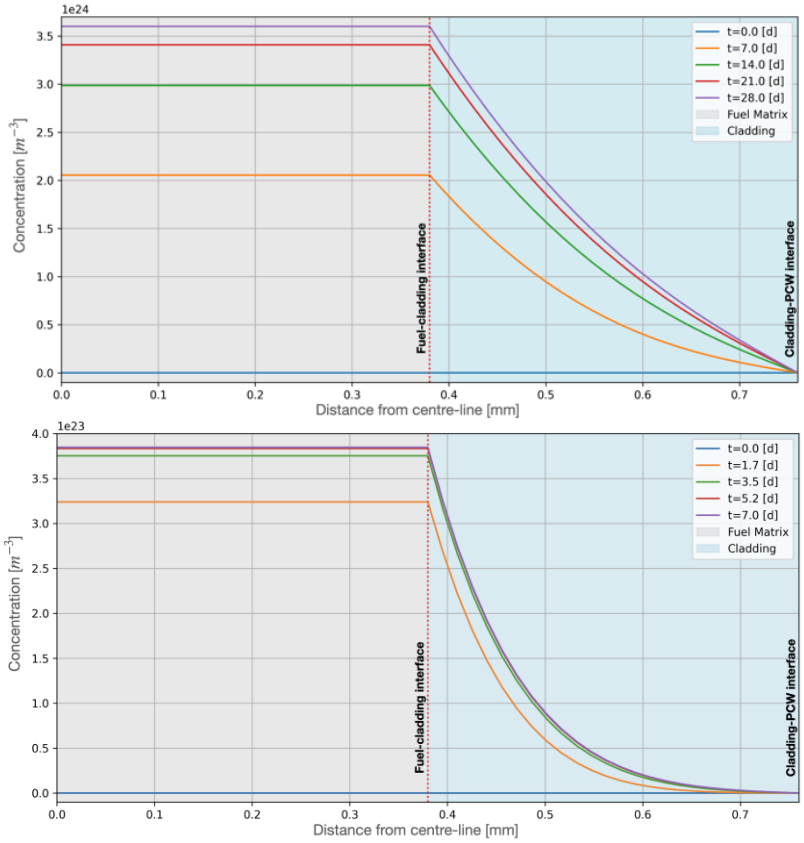

Figure 3. The concentration profile of Xe-133 (top) and Xe-135 (bottom) over fuel and cladding region as simulated by the Release Model.

Noble gases (such as Xe), are the most volatile within the fuel and cladding due to their gaseous state and chemical inertness [15]. Due to this, the derived Release Coefficient of Xe served as an upper limit for the release rates of other volatile fission products, enabling the prediction of what isotopes are not expected to be released under normal reactor operating conditions. An example of the Release Model's application is the simulated concentration profiles of Xe-133 and Xe-135 in the fuel and the cladding, which are illustrated in Figure 3.

3.3. Defining the Fuel Defect Indicators

By calculating the maximum release rates of volatile fission products based on the upper limit set by the noble gas Xe, the Release Model could identify Fuel Defect Indicators, i.e. what can only be released in case of a fuel defect. Three types of Fuel Defect Indicators have been defined:

1. **A change in the Xe-133/Xe-135 ratio**: Under normal operational conditions, a substantial portion of Xe-135, which possesses the shorter half-life of the two Xe isotopes, decays as it diffuses through the cladding, as illustrated in Figure 3 (bottom). However, in the presence of a fuel defect that compromises fuel cladding integrity, Xe is allowed to encounter water more promptly than it would with an intact cladding facilitating its earlier release. Consequently, a greater quantity of Xe is released into the water before it has the chance to decay. This is true for both Xe-133 and Xe-135, but due to the longer half-life of Xe-133, the effect is relatively less significant, leading to a change in the Xe-133/Xe-135 ratio in case of a fuel defect [11].
2. **Singular Fuel Defect Indicator radioisotopes**: The detection of singular radioisotopes that, according to the Release Model, could not have been released through diffusion processes in the cladding.
3. **Overall increase:** An overall increase in activity of the detected radioisotopes in the Volatile Fission Products group as defined for the Isotopic Baseline would also provide an indication of fuel defect.

3.4. Ageing Management

Ageing-related degradation of a reactor's structural components is of critical concern to nuclear installations, especially for an installation such as the HFR, which has been in operation for over 60 years. This necessitates ageing management, encompassing monitoring, prevention, and mitigation of degradation [16, 17].

The RMP is capable of long-term trending of radioisotope activity in the PCW, which enables the programme to track potential increases (or decreases) in certain radioisotope activities over an extended number of operational cycles. The radioisotopes relevant to the ageing of reactor components were defined as ‘Ageing Indicators’. The monitoring of such radioisotopes can give insight into the ongoing ageing processes within the reactor. To assess the GRS data related to Ageing Indicators, a robust linear model is employed to accommodate data variability and outliers.

An example of a potential Ageing Indicator for primarily aluminium based reactors, such as the HFR, is the activity of Na-24, which originates from the Al-27(n,α)Na-24 reaction. Assuming that the aluminium fuel cladding oxidation rate (i.e. of fresh fuel introduced during reactor stops) and PCW demineraliser filtration system are well characterised and stable over the period of evaluation, a long-term increase in Na-24 activity may thus find its origin in the increased corrosion rate of structural components in the primary circuit.

3.5. Combining in the Reactor Monitoring Programme

Developed in Python, the RMP combines the components outlined in this methodology. The RMP operates on a dynamic database of GRS measurements from 2022 and 2023, serving to calculate the Isotopic Baseline. This database is updated with each new GRS measurement that is within the Isotopic Baseline confidence interval, which in turn prompts the RMP's backend to revise the Isotopic Baseline with the updated dataset. Thus, the RMP is capable of handling ‘in-baseline’ evolutions of the radioisotope concentration. This is essential for incorporating variations in reactor operations and/or change and additions of structural materials and components over time, as well as including the effects of structural component ageing. The singular Fuel Defect Indicators are included in a Python list and the programme checks whether any have been detected in a certain week, as well as whether the Xe-133/Xe-135 ratio and Volatile Fission Products group are within baseline. The frontend, aimed at facilitating user interaction, provides a straightforward interface using Python widgets for the entry of new GRS measurements. The programme processes the substituted GRS data and provides statements about the operational state of the reactor as an output.

4. Results and Discussion

4.1. Derivation of the Isotopic Baseline

The Isotopic Baseline, derived from GRS data for specific groups, exhibits trends in the radiochemical composition of the PCW during reactor operational cycles that align with expectations pertaining to the behaviour of the given group. This is demonstrated by the Isotopic Baseline for the radioisotopes in the Structural Components group and Contaminant group in Figure 4. The accuracy of the Isotopic Baseline has been verified by training the model on GRS data of 2022 and the first half of 2023, and further tested against data from the latter half of 2023. This approach confirmed the model's reliability and confirmed that it could adapt to changes in radioisotope activity over time.

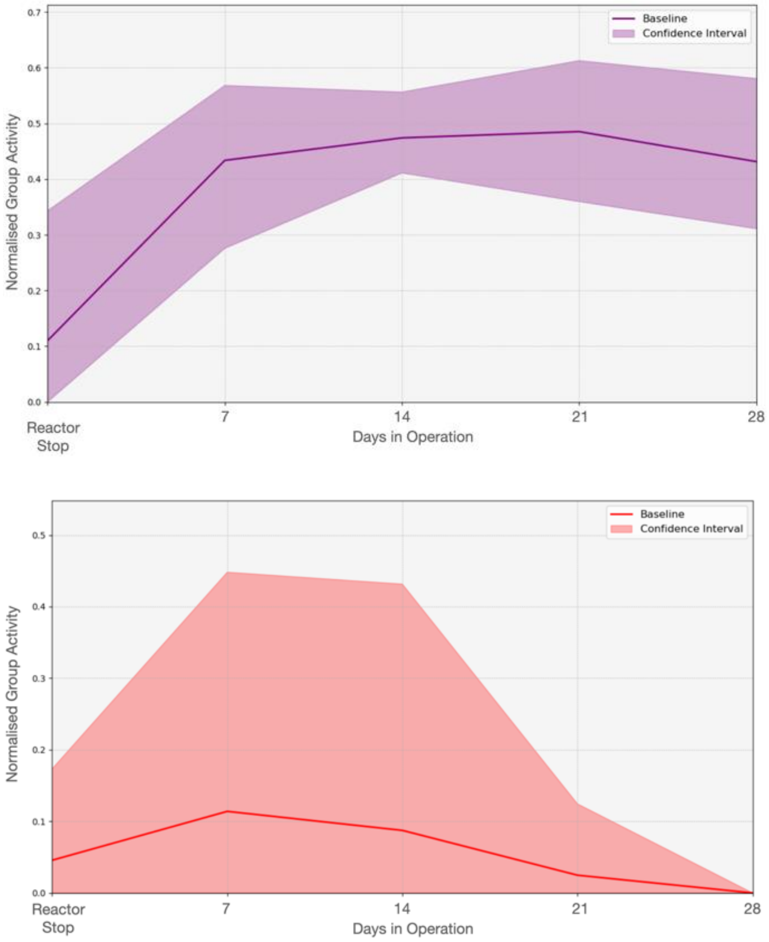

Figure 4: Isotopic Baseline profile for the Structural Component group (top) and Contaminant group (bottom). The days on the x-axis refer to the number of days in reactor operation, with ‘0’ referring to reactor stop.

For the Structural Components group, containing for example Na-24, Mn-54, and Cd-115 (Figure 4, top), the Isotopic Baseline exhibits a predictable increase in activity from structural elements in the PCW upon reactor start-up. This occurs as during reactor stops, dissolution rates of oxide films on structural components increases due to a shift in PCW chemistry, leading to a high concentration of structural component isotopes in the PCW upon reactor start-up, which subsequently undergoes neutron activation. A plateau level is reached around week two of reactor operation, as an equilibrium is reached between the production of radioisotopes belonging the Structural Components groups, radioactive decay of the produced isotopes and removal via the PCW demineraliser filtration system.

The Contaminants group (Figure 4, bottom), containing Sb-122, Sb-124 and Np-239, exhibits a distinct peak during the first week following the start of the reactor operational cycle, and thereafter reduces over the remaining reactor operational period. This trend is due to that source contaminants entering the PCW are of a finite quantity. Sb contaminants are introduced in the reactor stop periods during which the basin water mixes with the PCW, transporting Sb contaminants from the polymer materials in the basin to the PCW, and Np-239 is produced via the U-238(n,γ)U-239(β-decay)Np-239 reaction channel, whereby the U-238 is introduced into the PCW as tramp uranium on the fuel plates of fresh fuel loaded during the reactor stops.

Not visualised are the pH/Eh Related Products and the Volatile Fission Products. pH/Eh Related Products are only detected in stop as expected, and Volatile Fission Products keep increasing until the third week and then level off, indicating a steady state has been reached. The behaviour of the Volatile fission products was thus as expected and this was further validated by the Release Model, which can simulate the release of such isotopes from the fuel matrix through the cladding.

4.2. Performance of the Release Model

The Release Model's performance was validated by comparing noble gas activities simulated by the model to actual GRS data from the reactor's PCW during steady-state operation. The Release Coefficient established an upper limit for the release rates of volatile fission products. Rather than providing precise value estimations, the Release Coefficient serves to define a threshold, beyond which the presence of certain radioisotopes in the PCW indicates an increased release rate potentially caused by a fuel defect. The Release Model proved effective in identifying Fuel Defect Indicators. For example, it demonstrated that radioisotopes like Kr-88 should not be detectable under normal conditions, given the decay within the fuel matrix and cladding. Therefore, detection of such radioisotopes would suggest a fuel defect (e.g. cladding degradation, excessive porosity and/or cladding penetration).

4.3. Evaluation of the Fuel Defect Indicators

The Release Model could be used to define the following Fuel Defect Indicators:

- **The Xe-133/Xe-135 ratio**: A confidence interval was defined for the Xe-133/Xe-135 ratio for each week in reactor operation based on the available GRS data. This is visualised Figure 5, where the ratio evolves from 0.4 at day 7 of reactor operation, to 0.9 at day 28. This is due to that Xe-135 reaches steady state quickly, but the Xe-133 concentration keeps increasing over the reactor period. If the detected ratio is below the defined confidence interval (thus relatively more Xe-135 release), this can indicate a fuel defect.
- **Singular Fuel Defect Indicators**: Radioisotopes that should not be released through the fuel cladding (based on the upper limit of the Release Coefficient derived from xenon in normal steady-state operation), such as Kr-88, Sr-91, I-134 and Cs-138. Historic GRS data showed that these radioisotopes were indeed not detected during normal steady-state operation.
- **Complete group activity:** The whole ‘volatile’ group may be employed as the third Fuel Defect Indicator, as a significant activity increase of this group may be a result of reduced confinement of volatile radioisotopes present in the fuel matrix, through a fuel defect.

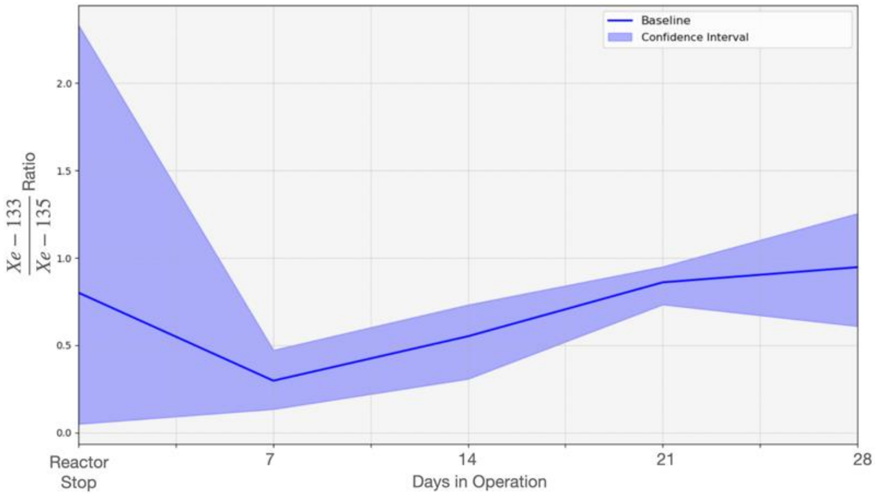

Figure 5: The Xe-133/Xe-135 ratio over the reactor operational period.

4.4. Ageing Management Insights

Analysis of the collected data over time revealed a steady increase in the activity of Ageing Indicators, suggesting an ongoing ageing process within the reactor's structural components. This trend appears to be linear in nature and could be correlated to a global increase in corrosion rate within the primary circuit. However, attributing the trend solely to corrosion rate increases lacks consideration for factors such a replacement of components/component material types and decreases in demineraliser filtration efficiency. This would require more detailed analysis, which is not conducted within the scope of this work. Figure 6 serves as an illustration of the RMPs ageing management function, depicting the activity pattern over time for Na-24.

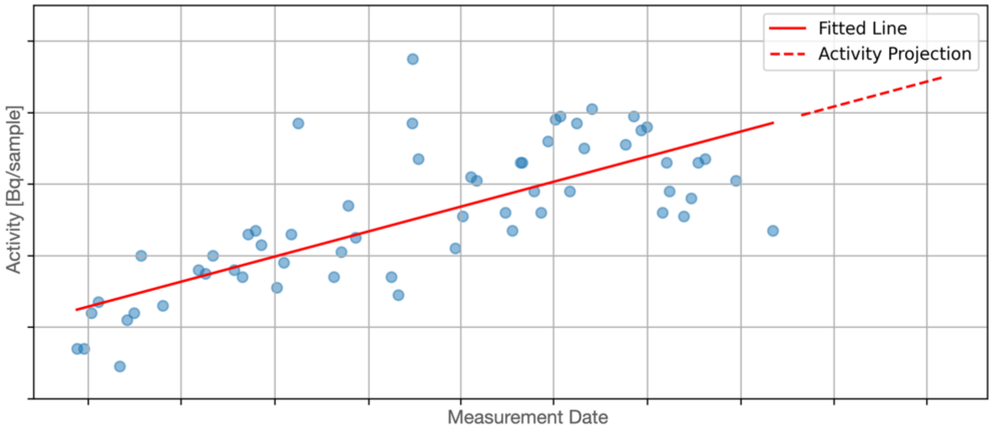

Figure 6: Increase in activity of the Ageing Indicator Na-24 over time. A fitted line from robust linear model shows a seemingly linear increase. Based on the fit an expectation for future activity can be plotted.

4.5. Integration and Adaptability of the Reactor Monitoring Programme

The RMP frontend consists of an interface using Python widgets to substitute all relevant data from a specific week's reactor operational period. This consists of sampling date & time, analysis date & time, the current operational week, and the GRS activity results for all radioisotopes. The RMP backend subsequently runs on the described methods, enabling precise and timely detection of operational deviations and potential fuel integrity issues. It might for example indicate that the activity of the radioisotopes in the 'Volatile Fission Products' group is within confidence interval of the Isotopic Baseline, but above average, or that a certain Fuel Defect Indicator is detected.

The RMP methodology described in this paper is adaptable to other LWR reactors. By analysing the GRS data of a reactor and grouping radioisotopes based on the reactor structural components and fuel type, the Isotopic Baseline can be customised for each reactor setting. The programme is easily adaptable to reactors using U_3Si_2-Al fuel with aluminium cladding, where the RMP's existing Fuel Defect Indicators apply directly. However, when different fuel or cladding materials are used, adjustments to the Release Model are necessary to accurately reflect the unique release behaviour of fission products and recalibrating the Fuel Defect Indicators accordingly. The approach to ageing management, which involves monitoring isotopic activity over time to detect structural degradation, is universally applicable and primed for further refinements.

5. Conclusion and Recommendations

The RMP is designed as a supporting tool for experts, facilitating the assessment of weekly in-line GRS data for nuclear reactors. By integrating three critical components: The Isotopic Baseline, the Release Model, and Fuel Defect Indicators, the RMP enhances the expert's ability to perform real-time evaluations of reactor operational states and early detection of fuel anomalies. The Isotopic Baseline uses historical GRS data to establish a reference point for normal operation levels, allowing for the immediate identification of deviations in isotopic activity. The Release Model, developed in Python, predicts fission product releases, supporting the identification of Fuel Defect Indicators. The Fuel Defect Indicators aid by quantifying specific changes in the GRS data that can signify anomalies. Through its systematic approach, the RMP transforms weekly GRS measurements into actionable insights, aiding in maintaining reactor safety and efficiency.

The adaptability of the RMP to different reactors with different fuel types or cladding materials has not been tested and represents a promising avenue for future research and application. Future enhancements of the RMP involve the incorporation of advanced algorithms to more accurately assess the origin of potential increases signalled by the ageing management component, distinguishing between ageing and reduced filter efficiency. Presently, the volume of GRS data is sufficient for defining a weekly Isotopic Baseline. With an increase in available data, the Isotopic Baseline can potentially be defined for shorter time intervals, such as for each day in operation, leading to more precise and representative analysis of new GRS data. More data would also lead to more accurate trends for Ageing Indicators, which can aid an analyst in ageing management.

6. References

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