Preventive Checking of Nuclear Fuel Rods

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Abstract. To ensure the safe operation of nuclear reactors, operators continuously monitor the operating parameters and adjust accordingly. This is aided by monitoring the primary coolant for the presence of fission products. This monitoring however only provides information about the complete reactor assembly. For information on the integrity of individual fuel elements, the "NIP-methode" (sipping method), as described in this article, can be used. Each fuel element is individually sipped, the obtained samples measured with a spectrometer, and the resulting spectra analysed for the presence of ^{60}Co (corrosion) and ^{137}Cs (breached/leaking). Using the NIP-methode the 30 fuel elements of the Hoger Onderwijs Reactor Delft are analysed. The method revealed a positive correlation between the presence of ^{60}Co in the sipped sample of a fuel element and a visual defect reported on the element.

KEYWORDS: Nuclear fuel elements, preventive checking, NIP, sipping tests, nuclear safety.

1   INTRODUCTION

The safety of nuclear reactors has always been of the utmost importance to protect both environment and operating staff from the potential risks involved with operating a reactor and working with fissile materials. Securing this safety is the task of the operating company of the nuclear reactor [1]. To this extent operators continuously monitor the operating parameters and adjust accordingly. In their work they are aided by the continuously monitoring of the release of fission products formed in off-gasses and pool water/primary coolant or in absence of such on-line systems periodical manual measurements are performed in accordance to regulations [2]. At the Hoger Onderwijs Reactor Delft (HOR) the off-gasses are continuously monitored while the pool water is manually measured twice a week. The dose load from the pool water is kept as low as reasonably achievable by using ion exchange resins. The main drawbacks of these (continuously) monitoring methods are that they only trigger a responds after fuel element integrity has been lost (i.e. increase in fission products detected) and they only give information about the complete fuel element assembly.

For testing the integrity of individual fuel elements several analysing techniques are available, each with their own advantages and drawbacks. Commonly described methods are visual inspection, eddy current, ultrasonic testing, gamma tomography, and sipping tests [2, 3]. These methods have in common that they are non-destructive, in contrast to leaching tests [4]. During visual inspection the fuel elements are closely examined by eye or camera for damages. However, this method only allows for inspection of the outer surface of the element [2]. Eddy current is used for the detection of cracks and oxide layer thickness on fuel rods [3]. Ultrasonic testing allows for detection of anomalies or water [3] within the fuel elements. Where eddy current testing uses an electric current [5], ultrasonic testing uses sound. However, the used probes must be adjusted to the shape of the fuel elements. Gamma tomography uses a collimated high energy resolution gamma spectrometer to detect the gammas emitted by the fuel elements. By imaging from multiple directions around the fuel element, 2D-slices can be reconstructed. This method is used to study the behaviour of the fission products within a fuel element, but can also be used for integrity analysis in safeguard. Gamma tomography can be both used for the whole assembly, as well as for individual fuel elements. Individual leaking rods within the fuel element can be identified. [6-8] Using gaseous fission products as marker, causes time constraints due to short half-lives as well as difficulty in identifying leaking rods from low-burn up rods due to low gaseous fission product formation [8]. Sipping tests, on the other hand, allow for the testing of complete fuel elements, and is ideal for determining whether or not a leak is present, or as to do a preselection for subsequent testing [3].

The NIP-method applied in the current research is based on the sipping-method reported by Perrotta et al. [9] in 1996. In their procedure individual fuel elements were subjected to individual sipping tests in open aluminium tubes. These tubes were situated in an open pool. The pool water was forced out of the aluminium tubes by injecting demi-water from the top of the tube. Such open design is prone to

contaminations. To avoid contaminations and increase accuracy Linge et al. [10] modified the design of the aluminium tube to a closed design, with the Demi-water for flushing the NIP-apparatus supplied via tubing to reduce contamination with pool water. The size of the apparatus was reduced to decrease the amount of (demi)-water present in the apparatus. These modifications decreased the detection limit of the measurements.

In the past the sipping method has been used to identify leaking fuel elements within nuclear fuel element assemblies [9, 11]. However, the method can also be used preventively, due to the detection of cobalt-60. In this work we show how our NIP-method can be used to identify (potentially) faulty fuel elements in a very early stage. As we are measuring leaked radioactivity with our NIP-method, even microscopically small leaks can be discovered which would not have been with found by visual inspection.

2   METHOD
2.1   Set –up
The design of the NIP-apparatus is based on the design by Perrotta et al. [12] and adapted by Linge [10]. The NIP-apparatus consist of a square shaped aluminium tube sealed at the bottom and with a lid at the top (Figure 1). Its dimensions are 10 cm by 10 cm by 100 cm. The lid is closed leak-tight after placement of a fuel element. The NIP-apparatus is situated in the reactor pool about 3.5 m below the water level (Figure 2), providing sufficient radiation shielding to allow safe transport of the fuel elements to the NIP-apparatus. The NIP can be flushed with Demi-water (Inhouse demi-water system) through the attached tubing to expel any primary cooling water. After flushing, the water in the NIP is circulated with a velocity of 4 m/s using a centrifugal pump.

Figure 1: Schematic representation of the NIP-apparatus [10].
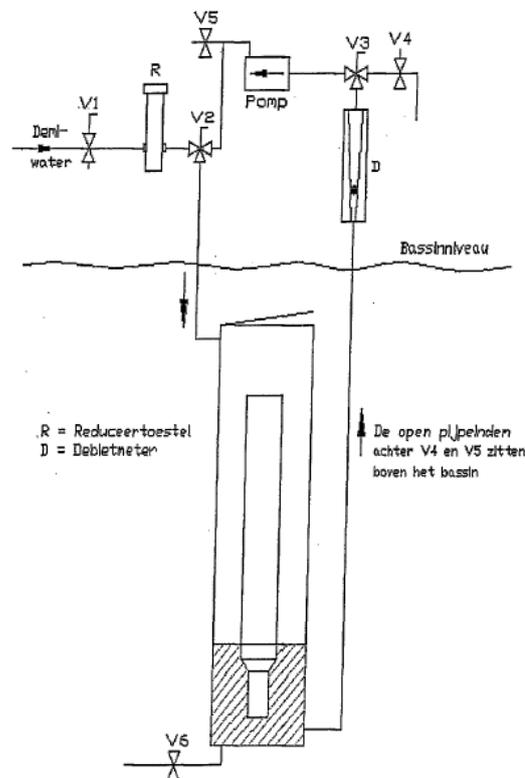
Figure 2: Position of the NIP-apparatus within the pool of the research reactor
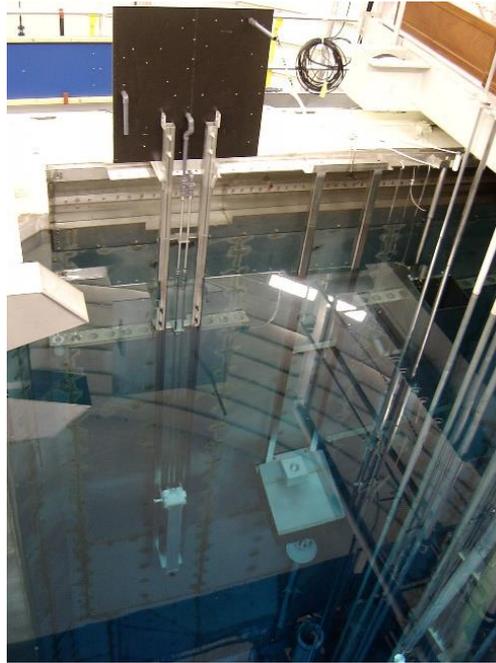
2.2   Sample generation

A reference sample of about 600 mL is taken immediately after flushing. The water in the NIP-apparatus is subsequently circulated for 3 to 7 days. At the end of this circulation period a final sample of about 600 mL is taken. During circulation extra samples can be taken.

2.3   Sample analysis

The samples are weighted to allow determination of the activity concentration after analysis.

All samples are measured for three days using a gamma spectrometer (NaI) connected to a PC with GammaVision software. Energy and efficiency calibration of the spectrometer is performed by using a certified ^{137}Cs source. Three regions of interest (ROI) are determined: 662 keV for ^{137}Cs, and 1173 keV and 1333 keV for ^{60}Co. The number of pulses measured within the ROIs are converted to an activity concentration. The spectra are also visually inspected for any peaks outside the ROIs that might require further investigation. The spectra are analysed using GammaVision software (GammaVision 6, Ortec). Additional information on the history of the fuel elements is requested if any anomalies are found. Some samples are also measured with a higher resolution spectrometer (HPGe, Canberra) connected to the same PC, and the obtained spectrum compared to spectra of pool water.

3   RESULTS AND DISCUSSION
3.1   Calibration of the spectrometer and determining the ROIs.

For accurate measurements of the generated samples the used gamma spectrometer needed to be calibrated. To this extent a certified ^{137}Cs source was measured for 18000 s, as well as a ^{60}Co source. The ^{137}Cs source had similar dimensions as the samples. The obtained spectra were used to perform an energy and efficiency calibration of the spectrometer. These spectra were also used to determine the regions of interest (ROIs). For the used spectrometer, the ^{137}Cs ROI was from channel 267 to 314 and the ^{60}Co ROIs were from channel 476 to 533 and channel 542 to 604. The efficiency of the gamma spectrometer was determined using equation 1, in which K_{eff} was the calculated efficiency coefficient in Bq/cnt, $A_{^{137}Cs}$ the activity of the used source at time of measurement in Bq, t the measurement time in s and $cnts_{ROI_{^{137}Cs}}$ the amount of measured counts in the corresponding ROI in CNT. For the spectrometer used in this research the efficiency is $K_{eff} = 183$ Bq/cnt. It must be noted that this is an underestimation of the efficiency at the ^{60}Co-ROI.

$$K_{eff} = \frac{A_{^{137}Cs} * t}{cnts_{ROI_{^{137}Cs}}} \qquad (1)$$

3.2   Sample generation

Due to the large amount of fuel elements (Table 1), long NIP times and sample measurement times the fuel elements were measured over a 12 month period. Furthermore, due to the long spectral measurements, acquired samples sometimes had to wait for measurement up to four weeks. As only long lived nuclides were chosen as key nuclides, this had no significant effect on the resulting spectra. Any very short lived radionuclides already had decayed before the fuel elements were handled, as at least a 24 h waiting period was taken into account between reactor shutdown and handling the fuel elements to ensure the safety of the operators handling the fuel elements.

Table 1: Fuel element statistics. Per fuel element at least two samples are measured: the reference and the final sample.

| | Fuel elements | | Total |
	Standard element	Control element	
Reactor assembly	16	4	20
In-pool storage	8	2	10
Measured samples	-	-	64

3.3   Obtained spectra

3.3.1   General observations

For all measured fuel elements it was noted that the NIP-sample showed an increase in overall activity concentration compared to the reference samples, even when no (significant) peaks were observed. When multiple samples were taken at different times during NIP circulation, an increase of activity in time could be observed. Even a NIP-measurement without a fuel element present showed a slight increase of activity in the sample compared to the reference measurement; Be it a smaller increase compared to measurements with a fuel element. This increase in overall activity can be attributed to seeping in of pool water into the NIP-apparatus. However, this seeping behaviour seems to occur randomly. To this extent additional long term NIP-measurements without a fuel element are conducted (Figure 3). Another explanation can be the diffusion of volatile products through the cladding of the fuel rods.

Figure 3: ^{60}Co activity concentration compared to the circulation time for NIP-measurements without fuel elements.
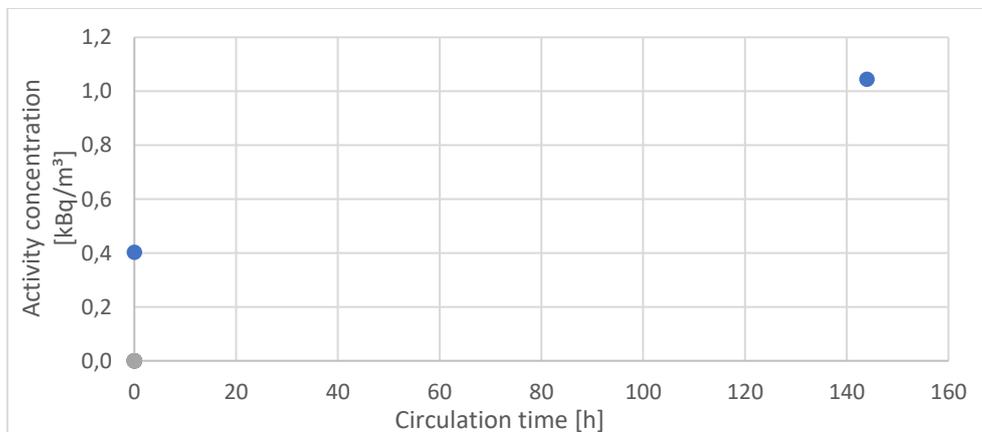
Table 2: Overview of the measured fuel elements.

	Reactor assembly		In-pool Storage		Total
	Standard element	Control element	Standard element	Control element	
Clean	9	2	3	2	16
^{60}Co contamination	7	2	4	0	13
^{137}Cs contamination	0	0	1	0	1

3.3.2   Clean spectra

Most of the sipped fuel elements provide ^{60}Co- and ^{137}Cs-free spectra (Table 2). No leaks or corrosion is found with these fuel elements. An example of clean spectra is provided in Figure 4. The reference sample and the sample both only show background activity. The peaks visible in the spectra outside of the ROIs are caused by natural background radiation and are still present when measuring without any sample present. (Data not shown.)

Figure 4:The spectra obtained from the several samples of a fuel element. The red bands mark the ROIs. A) reference sample, B) sample after 72 h of circulation.
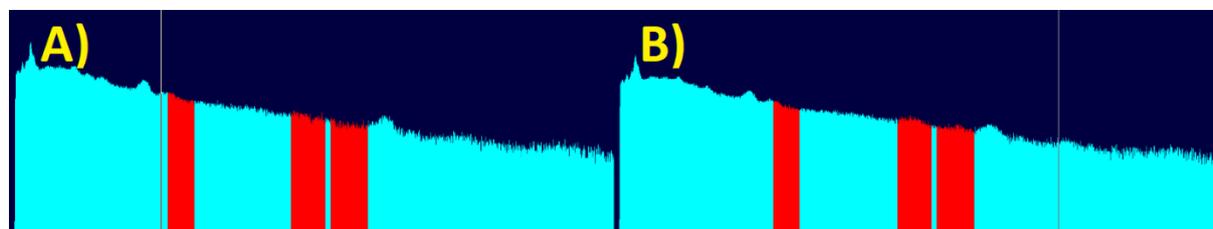
3.3.3 ^{60}Co containing spectra

Some of the measured fuel elements show a ^{60}Co contamination (Figure 5). ^{60}Co is most likely caused by activation of materials present in the reactor pool [13]. To determine whether the ^{60}Co contamination is dissolved in the water present in the NIP-apparatus or caused by flakes in the water, the sample was passed through a 0.45µm filter. Measuring the filtered NIP-sample (Figure 5 C), it can be seen that the ^{60}Co contamination is caused by dissolved ^{60}Co, indicating the contamination is most likely coming from the fuel element itself. One of the fuel elements also shows a ^{54}Mn contamination (Figure 6).

From several fuel elements the obtained NIP-sample is also measured on a HPGe spectrometer and the obtained spectrum compared to spectra obtained over time from the pool water. The ^{60}Co concentration in the pool water is estimated at 10 kBq/m³, based on the obtained pool water spectra. The obtained concentrations of ^{60}Co for the fuel elements that show contamination are usually higher, indicating that the ^{60}Co is originating from the fuel element itself.

The nuclear fuel element cladding is composed of an aluminium alloy. Even though nuclear grade materials are used, they are never 100% pure and slight contaminants can be present [14]. Known contaminates in alumina are for instance magnesium [15], manganese [16], cobalt, zinc and iron [14]. The contaminants, like the cladding itself, become activated during operation of the nuclear reactor. Whereas the cladding is designed to produce only short lived radionuclides, the contaminants can activate to long lived radionuclides [17]. Due to corrosion these (activated) contaminants can be released from the fuel element cladding.

To find a possible explanation for the detection of ^{60}Co the history of the fuel elements is examined. All kinds of parameters are considered, including age of the fuel element, operational time and position in the pool. None of these parameters provided an explanation. For some fuel elements it is reported that they have some sort of visual defect, for example a small scratch or pit. A positive correlation between ^{60}Co detected and a visual defect is found (Table 3): 0.38. The aluminium cladding is more likely to corrode when scratches or pits are present on the cladding [18], due to removal of the protective

aluminium oxide layer [15, 16]. Furthermore, the (local) pool water quality has an influence on the corrosion process of the cladding [2, 3, 18]

Figure 5: The obtained spectra from the samples of a fuel element contaminated with ^{60}Co. The red bands mark the ROIs. A) reference sample, B) sample after 96 h of circulation and C) same sample but filtered over a 0.45µm filter.
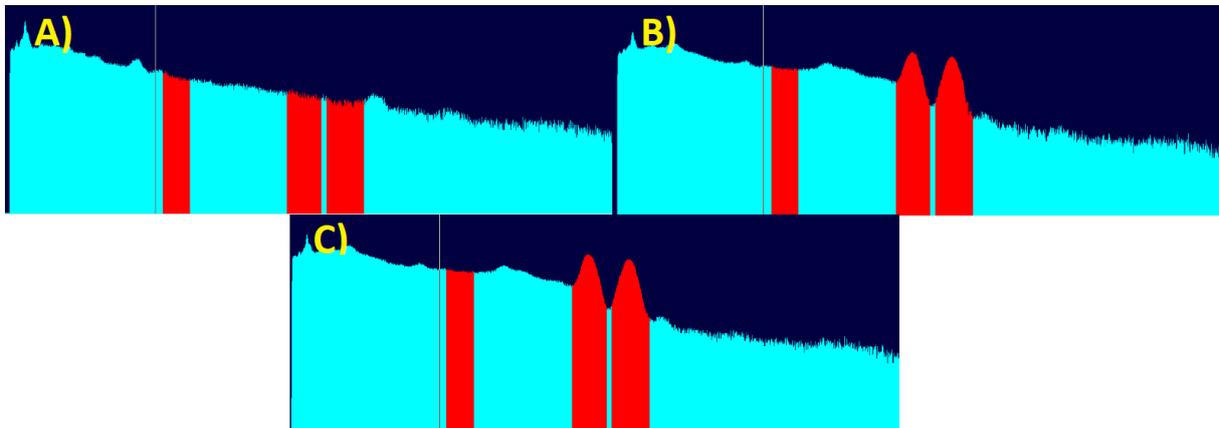
Table 3: Correlation between a visual defect found on a fuel element and the presence of ^{60}Co

		Visual defect		Total
		Yes	No	
^{60}Co	Yes	8	5	13
detected	No	4	13	17
	Total	12	18	30

Figure 6: The obtained spectra of a fuel element. Red bands mark the identified ROI. The arrow indicates the formation of a peak at approximately 834 keV, corresponding with ^{54}Mn. A) reference sample, B) sample after 68 h of circulation, C) sample after 144 h of circulation and D) sample after 237 h of circulation.
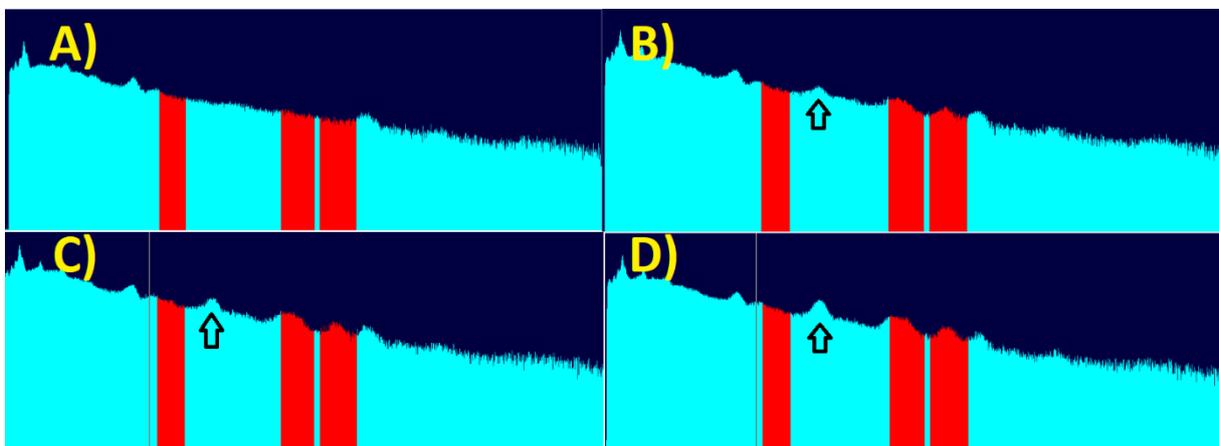
3.3.4 ^{137}Cs containing spectra

The presence of the isotope ^{137}Cs is used to identify whether a fuel element is leaking. ^{137}Cs is a fission product with a long half live (30 years) [17]. Of course there are more isotopes formed during fission [13], however, ^{137}Cs is solely formed by nuclear fission and therefore allows for distinction between leaking of the fuel element and contaminations resulting from neutron activated research samples. Furthermore, the long half live of ^{137}Cs allows for identification of leaks in spent nuclear fuel elements even after long-term storage. Based on the spectra shown in Figure 7 a leaking fuel element is clearly identified. A clear peak is visible at the ROI of ^{137}Cs (Figure 7B) and the release of ^{137}Cs is estimated at 2.8 kBq/h. The fuel element was subsequently removed from the assembly and stored safely. It must be noted that the fuel element was never considered leak, as legal limits were never exceeded.

Figure 7: The obtained spectra of a fuel element with ^{137}Cs contamination. The red bands represent the identified ROIs. A) reference spectrum and B) spectrum after 3 days of circulation.
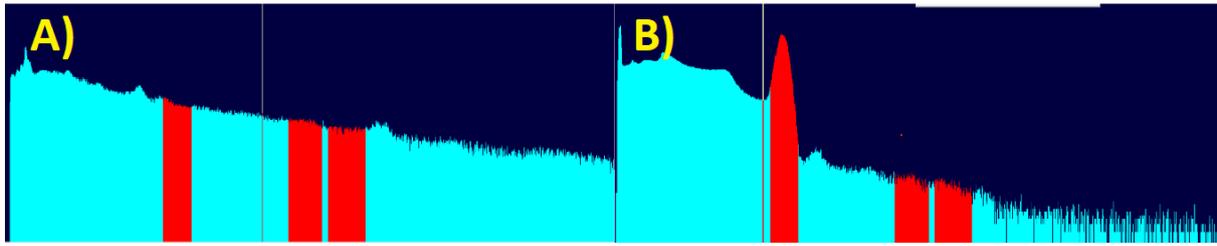
4   CONCLUSION

Even though the NIP-procedure can be time consuming, it is a straight forward method to identify corroding and / or leaking fuel elements. The presence of ^{60}Co in the NIP-samples indicated corrosion, while the presence of ^{137}Cs indicated a leaking fuel element. When ^{60}Co is found in the sample most of the time this co-insides with a visual defect on the fuel element.

5   ACKNOWLEDGEMENTS

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