

Experience with different methods for on- and off-line detection of small releases of fission products from fuel elements at the HOR

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Abstract. The paper gives an overview of the different methods for on-line and off-line detection for small amounts of activity used at the Hoger Onderwijs Reactor (HOR). The different installed techniques with their pro's and con's are discussed in more detail. During the HEU to LEU fuel conversion we encountered five cases in which the fission product concentration in the reactor pool water increased significantly. Although this increase never caused any exceeding of intervention levels as stated in our permit and safety analysis, the ALARA principle urged us to analyse the situation in more detail. As a result three new instruments were developed. The gained experience with all three new instruments is given. One of the new instruments, the 'RID-cascade', is described in more detail.

1. Introduction

The Reactor Institute Delft (RID) is part of the Faculty of Applied Physics which belongs to the Delft University of Technology. The RID is the Dutch national centre for multidisciplinary research and education involving the Hoger Onderwijs Reactor (HOR), nuclear radiation and radionuclides. Until 2005 the institute was an interfaculty facility. Since 2005 the interfaculty facility is split in a new research department 'Radiation, Radionuclides and Reactors' (R³) and the Reactor Institute Delft. Together they form the national focal point of expertise in the fields of reactor physics, neutron and positron radiation (including detection) as well as radiochemistry. At the heart of the reactor institute is the HOR as a source of neutron, positrons and radioisotopes. The HOR is a pool type research reactor operated 24 hours per day, 5 days a week at a thermal power level of 2 MW, attaining a modest thermal flux of about 2×10^{13} n/cm²/s. The core was compacted to 20 MTR-type LEU fuel assemblies during the HEU to LEU conversion which started in 1998 and was completed successfully in 2005. The 20 fuel assemblies are surrounded by beryllium reflector elements at three sides.

During the HEU to LEU fuel conversion of the HOR we encountered four cases in which the fission product concentration in the pool water increased significantly. Although this increase never caused any exceeding of intervention levels as they are mentioned in our permit and safety analyses, the ALARA principle urged us to analyse the situation in more detail. All cases have been related to a failure of the cladding of five elements respectively, one HEU fuel element and four LEU elements from the first batch of LEU fuel. Once an element has been identified to have a defect, it was removed from operation. The HEU fuel element defect and the first two LEU fuel element cases are described in [1]. These defects and the ALARA principle urged us to look for methods to detect small defects in fuel assemblies as fast and reliable as possible. We choose to detect these small defects by increasing the ability to detect small increases in activity fast and reliable within the reactor pool and the reactor building.

The following sections will give an overview of the already available detection methods and new developed ones, the gained experience with the different detection methods, a more detailed description of one of the new developed systems and conclusions drawn from the practical experience.

2. Overview of different on- and off line detection methods for small amounts of activity

Due to the needed sensitivity it is important to make a distinction between detection techniques for measuring small amount of activity and techniques for measuring high amount of activity. Both kinds of detection systems are available at the HOR but in this paper the focus is on detection methods for small amounts of activity. In practice different detection techniques are in place for on- and off-line determination of (increased) small amounts of activity within the reactor pool and containment, such as:

- Direct measurement of fission product activity in the reactor pool. This system makes use of a small ion exchanger system. Pool water from a depth of approximately 3 m is being led across a small anion and cation exchanger system. Underneath the anion exchanger a NaI crystal is placed for the detection of I-135. In this way the activity of all deposits in the anion exchanger are integral measured and with a high Na-24 background from the pool water. Because of this the sensitivity for the direct measurement of small amounts of increased activity due to fuel element defects appears to be low. This channel was original developed for natural convection mode of the HOR with upwards cooling flow instead of the downwards flow used with forced convection.
- Measuring primary cooling water delayed neutron activity. This system employs six BF₃ detectors counting delayed neutrons from fission products and is placed on a location shielded from gamma radiation in the primary cooling water circuit. In the case of the fuel element defects we detected with this measurement channel no increase of the signal.
- Direct measurement of air-borne activity concentration just above the reactor pool. This signal is referred to as ‘dust activity reactor building channel’ or ‘air-borne activity channel’. The dust activity monitor measures the deposit aerosol activity on a carbon filter using a G-M tube. This carbon filter is constantly fed by air directly from a suction point above the reactor pool. In this way we measure the decay products from the noble gasses Kr-88 (Rb-88), Kr-89 (Rb-89) and Xe-138 (Cs-138) released from the reactor pool, where as soluble activity like Na-24 remains in the reactor pool. This dust activity monitor proved that it is able to detect small amounts of activity due to fuel element defects. Due to its basic technical design, a G-M tube pulse counting system, it proved to be a reliable instrument over many years without a much need for maintenance. However the signal of the dust activity monitor can be masked by the hydrodynamics of the reactor pool.
- Direct measurement of (Ar-41) activity in the exhaust ventilation air stream. This gas activity channel is based on a scintillation detector behind a set of absolute filters and did not show an increase in indication in the case of the fuel element defects.
- Contamination check at the exit of the reactor building. At the two exits of the reactor building are hand and feet monitors positioned. They both detect alpha and beta radiation with a high sensitivity. In one of the five cases a contamination could be detected.
- Off-line analyses of ion exchanger beds from the fission product activity measurement system. The anion and cation exchanger beds of this system are weekly renewed and analysed. This proved to be a reliable indicator but the measured values depend on the power of the reactor, the amount of operation time and the effectiveness of the pool ion exchange system. . An other disadvantage is that it takes one week to get and indication.

- Off-line analyses of reactor pool water at the end of a production week. A sample of the reactor pool water from a depth of approximately 3 m is taken at the end of a production week and analysed for I-131 and I-133 after one day for the decay of Na-24. This also proved to be a reliable indicator but it suffers as with off-line ion exchanger bed analysis from dependence on the power of the reactor, the amount of operation time and the effectiveness of the pool ion exchange system.

During the period of having the fuel element defects and confusing data from the different detection techniques we developed the following instruments in addition to above listed to obtain more direct indicators of the reactor core performance:

- (1) A model to relate the pool water activity with the mean core fission products 'release constant'. This is explained in detail in section 4.
- (2) A wet sipping device to determine the fission product 'leak rate' of a separated, isolated element at suspense [2]. Furthermore the idea was to set a reference level for every element and to detect minor anomalies in the 'leak rate' at an early stage by regularly re-measure it during the lifetime. Disadvantages of the wet sipping device are that it is a time consuming measurement and that fuel elements are measured 'cold' which does influence the 'leak rate'. In the case of the five fuel elements with a defect we had no reference levels yet, but compared with elements with no defect an increase in release of activity could be demonstrated.
- (3) Understanding the different factors from the 'release constant' model and combining it with the merits of the above mentioned direct air system led to the development of a new device the 'RID-cascade', which measures on-line the air-borne activity concentration as it builds up from a direct coolant 'core cooling water'-air interface. Important aspects of the device are: high efficiency for fission or decay products, easy maintainability and easy to operate. This new measurement channel is described in more detail in section 6.

3. Experience with the detection of small releases of fission products

Experience with the different measuring methods concerning small releases of fission products was gained mainly from 2001 until June 2005. All events occurred during the HEU to LEU fuel conversion of the HOR. The first event around June 1997 could be deduced to a HEU element (D-12) which showed clearly pitting corrosion on an outer fuel plate [1]. The four other cases relate to LEU fuel elements from the first batch and occurred respectively around September 2001 (element E-06), April 2003 (element E-07), October 2003 (element E-03) and June 2005 (element E-05). Because the HEU element defect is a separate event and the time in between this event and the LEU cases is relative long, the experience with the detection of small releases of fission products is especially gained during the LEU cases.

The average value of I-131 activity in the reactor pool at the end of a production week is 20 kBq/m³ which is the same as the theoretical value based on a uranium contamination of the aluminium surface of the manufactured fuel elements of 0.01 µg/dm². The manufacturer guarantees a maximum uranium surface contamination of 0.1 µg/dm². During the first event the I-131 activity measured during the off-line analyses of reactor pool water increased slowly from 20 kBq/m³ to 140 kBq/m³. On basis of these findings it was decided to start a fuel element exchange program resulting in the exclusion of element D-12.

During the second event, which was the first with a LEU element, only the off-line analysis of reactor pool water showed a continuous but very slowly increase in I-131 activity to 100 kBq/m³ from September 2001 onwards until May 2002. In this period we developed the 'release constant' model, which allowed us in May 2002 to remove the defected LEU element (E-06) from the core.

During the following event around April 2003 the first indication of a possible element with a defect was observed by an increase in the 'dust activity'. As this was not seen in the previous cases, the

increase in ‘dust activity’ was not immediately connected to a possible element with a defect. When a minor contamination was detected on the floor of the reactor building and the activity of the dust monitor increased suddenly until 400 kBq/m³ it was decided to start a fuel element exchange program. LEU fuel element E-07 showed a defect and was removed from the core. In the week of the fast increase of the ‘dust activity’ also the ‘release constant’ increased significantly.

During the following event around October 2003 no increase in ‘dust activity’ was seen. Only an increase was seen in the ‘release constant’. After extensive visual inspection of all elements a defect was found on LEU fuel element E-3.

The until now last event occurred around June 2005 when a sudden increase in ‘dust activity’ was detected. After a core exchange program and extensive visual inspections of all fuel elements in the core a defect was found on LEU fuel element E-05.

Figure 1 shows the calculated release constant and the maximum ‘dust activity’ values for the four LEU fuel element defect cases in a production week. It can be seen that in the E-06 and E-03 LEU fuel element cases the ‘dust activity’ measurement channel gives a ‘false negative’ value. This ‘false negative’ indications did take place during autumn and winter. The reason for this is that during these colder periods the natural hot water layer is maintained more stable in the reactor pool. After having gained experience in detecting small activity increases, analyzing this winter / summer phenomena and because of the fast indication of the ‘dust activity’ monitor an improved ‘dust activity’ channel was designed and built. The following section will describe this new channel in more detail.

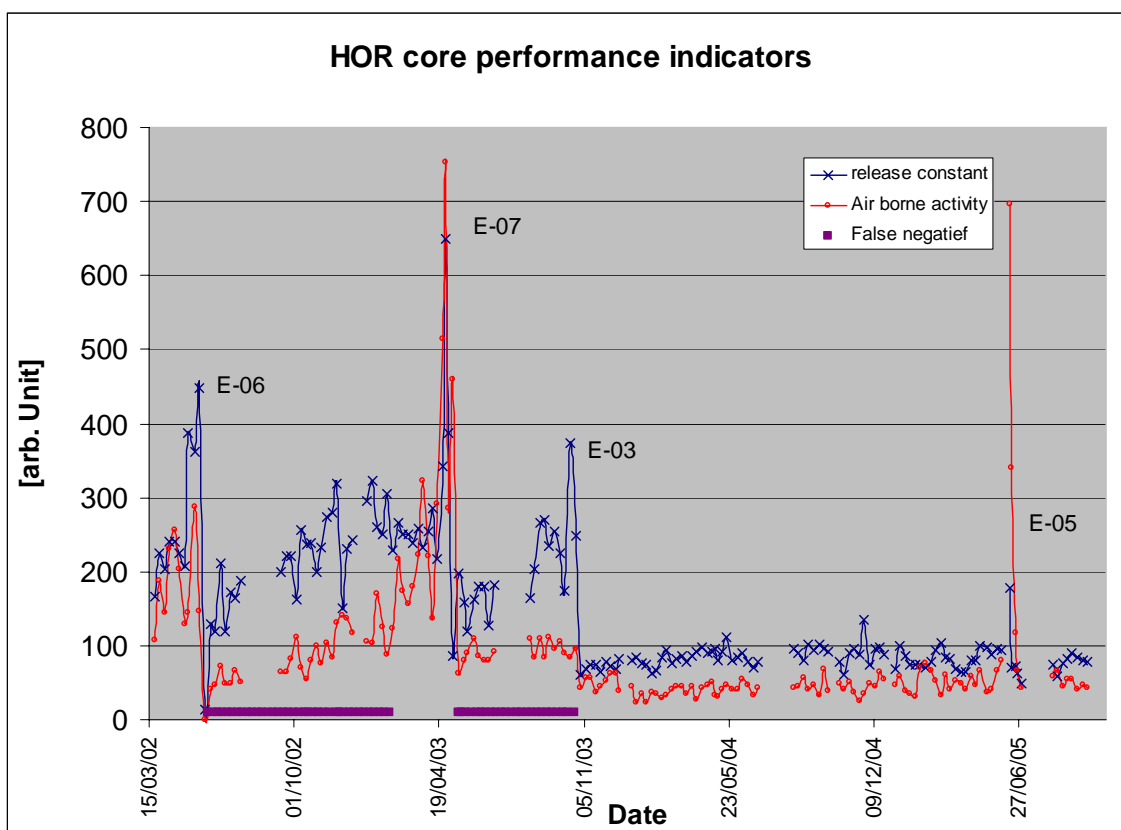


FIG. 1. Release constant and dust activity for all LEU element defects

4. Core fission products release constant

As mentioned we noticed from autumn 2001 a slow overall increase over several months of our regular I-131 pool water analysis at the end of the weekly reactor operation. However the signal suffered of great fluctuations dependent of the actual reactor performance.

To overcome this we introduced extra weekly pool water samplings at the beginning of reactor operation. As marker in the analysis we use the activity concentration of I-131 in the reactor pool water.

If the reactor at operation releases I-131 activity at a constant rate P (i.e. the release constant), than at infinite reactor operation time the pool activity will reach a saturation value A_s :

$$A_s = \frac{\lambda_I}{\lambda} \times P \quad (1)$$

where

λ_I is the physical decay constant of I-131

$\lambda = \lambda_I + \lambda_{\text{pool}}$ ion exchanger; i.e. the combination of λ_I and a constant introduced by the pool ion exchange system which constantly cleans the reactor pool water.

P is the release constant

With this assumption and with first order dynamics the activity during operation at time t will be:

$$A_t = A_0 e^{-\lambda t} + A_s (1 - e^{-\lambda t}) \quad (2)$$

where A_0 is the reactor pool activity at t is 0

During the weekend, where the reactor is shut down, the pool activity follows:

$$A_t = A_0 e^{-\lambda t} \quad (3)$$

with the same λ as the pool ion exchange system also runs through the weekend

By taking a reactor pool water sample after the weekend before start-up of the HOR and to compare this with the measured value of reactor pool water at the end of the previous production week, a half-life value of the pool ion exchange system is obtained. Combining this with the with the half-life of I-131 and the measured half-life value of the pool ion exchange a saturation activity value for I-131 can be obtained. From this saturation activity value the 'release constant' is calculated as a performance indicator which can be easily compared. This approach turns out to be a sensitive and reliable indicator even at very short reactor operation times as a few hours (see also figure 1).

5. Wet sipping device

This device consists of an in-pool container in which a fuel element at suspension is isolated from the reactor pool water. Once the container is closed the remaining pool water in the container is removed by flushing it with demineralised water of about ten times its volume. Hereafter the container with element is included in a closed water loop which can be sampled at different times. The activity of these samples analysed with a NaI detector as a function of the sample time is then taken to be an indicator of the failure of the element cladding.

On the next page in figure 2 and 3 respectively we show the results for the discussed elements E06 and E03. Even with this long sample times, no relevant activity could be detected at fuel elements which are not at suspect.

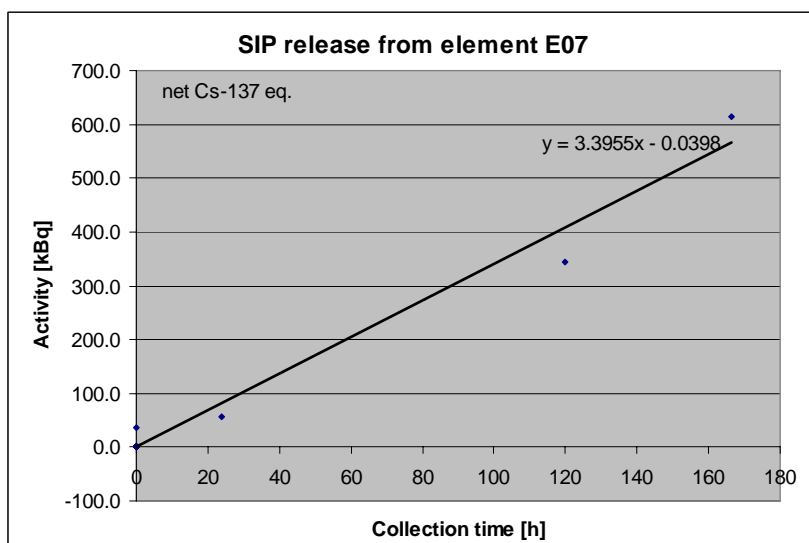


FIG. 2. SIP release from element E-07

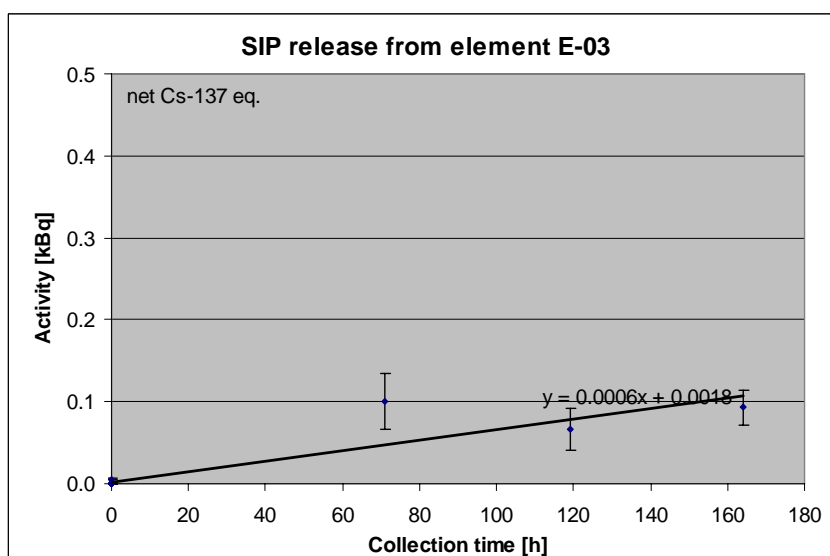


FIG. 3. SIP release from element E-03

6. The RID-cascade

The boundary conditions for the design of the new monitor were the following:

- (1) The signal should not be influenced by the natural occurring 'hot water' layer;
- (2) The sensitivity for small fuel defects should be as high as possible;
- (3) The influence of background activity like Na-24 from the pool water should be as low as possible;
- (4) Maintenance and calibration time should be as low as possible;
- (5) Simple interpretation and analysis;
- (6) Reliable over time.

Taking the above boundary conditions in consideration the RID-cascade was developed. Figure 4 gives an overview of this new monitor.

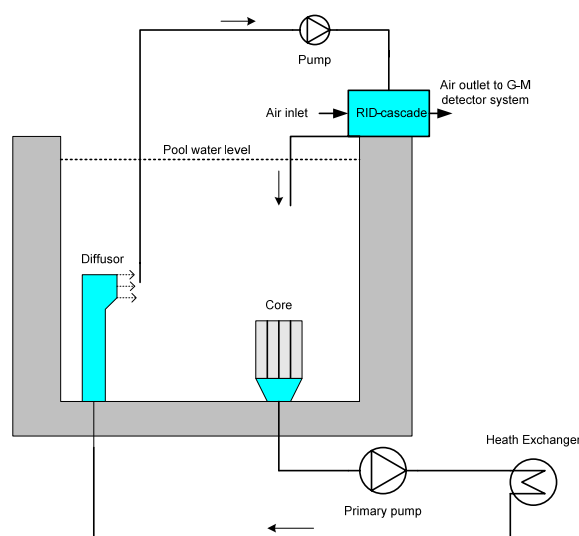


FIG. 4. Schematic overview RID-cascade

Boundary conditions (1) and (2) are met by sampling core cooling water from the heat exchanger. As can be seen in figure 4 this was realized by continuously sampling returning primary cooling water from the so called diffuser. Boundary conditions (3) and (5) and the gained experience with the 'dust activity' monitor shows that a direct coolant 'core cooling water'-air interface is needed. By choosing for a G-M tube counting system (figure 5) instead of gamma-spectroscopy using a Ge detector system boundary conditions (4) and (6) are met. The resulting design of the 'RID-cascade' is shown in figure 6 on the next page.

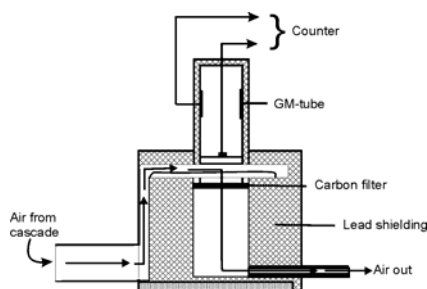


FIG. 5. Cross-section G-M detector RID-cascade



FIG. 6. The RID-cascade

The cascade on which the sampled core primary cooling water enters from the top can be seen clearly in figure 5. The air inlet of the RID-cascade is next to the air inlet of the ‘dust activity’ monitor. Figure 7 shows the indications of the RID-cascade (AHO2) and the ‘dust activity’ monitor (AHOR) during the start-up of the HOR. The RID-cascade signal follows the reactor power instantly and the signal is less prone to fluctuations compared to the signal from the ‘dust activity’ monitor. As can be expected the end value after stabilization of the RID-cascade signal is higher: 1000 kBq/m³ compared to 33 kBq/m³ from the ‘dust activity’ monitor. Because the air inlets of both systems are next to each other it is possible to compare the signals when the core cooling water sampling to the RID-cascade is stopped. In this case both systems show the same indication.

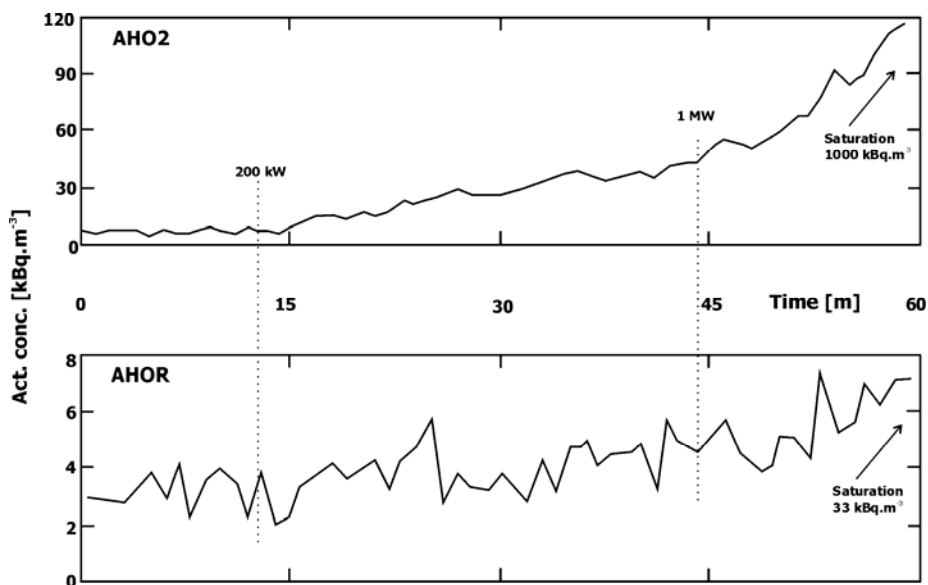


FIG. 7. Measurements results dust activity monitor and RID-cascade

7. Conclusions

During the period of having LEU fuel element defects, experience is gained in the detection of small releases of fission products. During this period three new instruments were developed: a 'release constant' model, a wet sipping device' and the RID-cascade. The 'release constant' model made it possible to detect the third LEU fuel element defect in an early stage. With the new RID-cascade a measurement channel is developed which is the result of all gained experience.

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- [2] PERROTTA, J.A., et al., Experience on wet storage spent fuel sipping at IEA-R1 Brazilian research reactor, *Annals of Nuclear Energy*, Vol. **25** (1998) 237-258.