

# Cesium and Iodine release fractions in a loss-of-coolant accident (LOCA)

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**Abstract.** A loss-of-coolant accident (LOCA) implies considerable damage to the core of a nuclear reactor and consequently the release of radioactive fission products. The safety analysis of a LOCA is defined in the various countries' regulations, e.g. as outlined in Regulatory Guide 1.183 (U.S. Nuclear Regulatory Commission, 2000). In this guide, a fuel rod gap inventory of 5 % of both the total <sup>131</sup>I and <sup>137</sup>Cs fuel inventory is assumed to be available for instantaneous release. A further 25 % (BWR) or 35 % (PWR) of <sup>131</sup>I, as well as 20 % (BWR) or 25 % (PWR) of <sup>137</sup>Cs, is assumed to be released as the accident progresses. These release fractions are valid for LWR fuel with a burnup up to 62 MWd/kgU. The Halden LOCA test series addresses a number of issues associated with the behaviour of high burnup fuel in LOCA conditions. As fuel segments with very high burnup (80-90 MWd/kgU) are employed in some of the experiments, it is considered of interest to investigate the release of <sup>131</sup>I and <sup>137</sup>Cs as one of the objectives. As a consequence the following examinations are carried out as part of each test:

1. Estimate of activity inventory of fuel pre-LOCA using the Oak Ridge National Laboratory (USA) software Origen.
2. Determination of activity content in effluent loop water by gamma spectral analysis.
3. Determination of contamination deposited within loop piping by gamma spectral analysis.
4. Determination of post-LOCA <sup>131</sup>I and <sup>137</sup>Cs inventory in rod by gamma scanning.

The gamma scan analysis of the fuel itself has proved challenging, depending on the severity of the fuel relocation. Nevertheless, although these results are affected by uncertainties, they point to lower <sup>131</sup>I release for the fragmented high burnup fuel commonly employed in the experiments than specified by NRC.

**KEYWORDS:** *Loss-of-Coolant accident, release, gamma spectroscopy*

## 1. INTRODUCTION

A loss-of-coolant accident (LOCA) implies considerable damage to the core of a nuclear reactor and consequently the release of radioactive fission products. The events at the Fukushima-Daiichi facility in Japan in March 2011 resulted in the release of fission products like the noble gasses and the volatile Iodine and Cesium. The safety analysis of a LOCA is defined in the various countries' regulations, e.g. as outlined in Regulatory Guide 1.183 (2000) issued by the U.S. Nuclear Regulatory Commission. In this guide, a fuel rod gap inventory of 5 % of both the total  $^{131}\text{I}$  and  $^{137}\text{Cs}$  fuel inventory is assumed to be available for instantaneous release. A further 25 % (BWR) or 35 % (PWR) of  $^{131}\text{I}$ , as well as 20 % (BWR) or 25 % (PWR) of  $^{137}\text{Cs}$ , is assumed to be released as the accident progresses. These release fractions are valid for LWR fuel with a burnup up to 62 MWd/kgU. The current safety criteria for loss-of-coolant accidents (LOCA) are based on experiments in the 1970s, which were largely conducted with fresh fuel (G.Hache, 2001). Changes in fuel design, introduction of new cladding materials and moves to high burnup fuel have generated a need to re-examine safety criteria and their continued validity.

## 2. THE HALDEN LOCA TEST SERIES

The Halden Reactor Project (HRP) LOCA test series addresses a number of issues associated with the behaviour of high burnup fuel in LOCA conditions. The first LOCA experiment in the Halden Boiling Water Reactor (HBWR) was carried out in May 2003 as an integral in-pile test on fuel behaviour under simulated LOCA conditions (Lestinen, 2004). The 12<sup>th</sup> test in this series was carried out in the spring of 2011. As fuel segments with very high burnup (80-90 MWd/kgU) are employed in some of the experiments, it is considered of interest to investigate the release of  $^{131}\text{I}$  and  $^{137}\text{Cs}$  as one of the objectives. Investigations concerning the release of  $^{131}\text{I}$  and  $^{137}\text{Cs}$  have been included as one of the objectives in tests 9, 10, 11 and 12, although the analysis of test nr.12 data is still work in progress. It is expected that these investigations will be continued and refined in future LOCA tests.

### 2.1 A SCHEMATIC OVERVIEW OF THE EXPERIMENTAL LOOP

In the LOCA test series a test fuel rod is mounted within a pressure flask in a test rig, in-pile and connected with a closed experimental loop, allowing for a controlled fuel failure. The entire loop is confined to the reactor containment, operated with heavy water, high pressure and forced circulation. A blow-down line from the pressure flask leads to a shielded dump tank with purification system and two water capture cells. Water samples can be collected from the cells when access to the reactor containment is allowed. A gamma monitor is positioned along the blow-down line and will observe radioactive material passing to the dump tank.

### 2.2 DESCRIPTION OF TEST PROCEDURE

The preparatory phase of the test involves steady state operation with power and pressure adjustments. The test is initiated by a main loop bypass, and the coolant water in the sealed off pressure flask is operated with natural circulation. Subsequently a blow-down to the dump tank removes more or less all of the coolant. With rapidly increasing temperature, the fuel rod failure is forced within minutes. Small amounts of steam are injected for the cladding oxidation. The test typically lasts about 5 minutes, being terminated with a deliberate reactor scram. Finally the pressure flask is filled with helium to secure dry storage. A significant amount of data, such as fuel temperature, rod elongation, rod pressure and loop radioactivity levels to mention a few, are monitored real time and kept for analysis. Gamma scanning examination of the pressure flask is carried out in the laboratories at the HBWR facility, and full PIE is carried out at the HRP hot-cell facilities at Kjeller.

### 3. EXAMINATION OF $^{137}\text{Cs}$ AND $^{131}\text{I}$ RELEASE

In a LOCA test with fuel failure radioactive material may have been released to either the pressure flask in the test rig, or the blow-down line and dump tank. In order to examine the release of  $^{137}\text{Cs}$  and  $^{131}\text{I}$  in tests nr.9, 10 and 11 the following examinations have been carried out:

1. Estimate of fuel inventory, using the Oak Ridge National Laboratory (ORNL) software Origen.
2. Determination of activity content in effluent loop water by gamma spectral analysis.
3. Determination of deposited contamination within blow-down line by gamma spectral analysis.
4. Determination of post-LOCA  $^{131}\text{I}$  and  $^{137}\text{Cs}$  inventory in rod using gamma scanning techniques.

The listed examinations draw a complete picture of the extent of the release. The  $^{137}\text{Cs}$  and  $^{131}\text{I}$  released and confined by the test rig is not measured, yet can be assessed based on the Origen fuel inventory estimate, the post-LOCA inventory, as well as the activity in the blow-down line and dump tank.

#### 3.1 CHARACTERISTICS OF TEST RODS NR.9, 10 and 11

Before the Halden experiment, the fuel in test nr.9 had a substantial burnup of 90 MWd/kgU due to commercial irradiation (Chomont, 2009). During the test, the rod experienced extensive fuel relocation and fragmentation involving the entire fuel stack, except for two top end pellets which appeared intact. About ¼ of the fuel stack was missing and a clad balloon in the lower half had twice the original diameter. The pre-test burnup of the fuel in test nr.10 and nr.11 was less than for test nr.9, yet still relatively high, at 61 MWd/kgU and 56 MWd/kgU respectively (Lavoil, 2010; OECD Halden Reactor Project, 2011). In both tests, the fuel rod experienced ballooning and burst of the cladding, however only limited fuel relocation.

#### 3.2 ESTIMATE OF FUEL ACTIVITY INVENTORY

The fuel inventory at the moment of LOCA is estimated for each test using Origen software. The inventory of  $^{131}\text{I}$  is estimated based on the isotopic composition upon arrival at the Halden Reactor facility, measured LOCA-test power history and a Halden Boiling Water Reactor adapted nuclide library. The inventory of the long-lived  $^{137}\text{Cs}$  is estimated based on the initial isotopic composition and enrichment, commercial operating history and burnup, as well as a corresponding nuclide library. The irradiation period in HBWR, typically lasting weeks or a few months, is also included; however the contribution to the total burnup and  $^{137}\text{Cs}$  inventory of the fuel is usually minimal.

#### 3.3 ACTIVITY IN EFFLUENT WATER AND DEPOSITION

Gamma analysis examinations of deposition in the blow-down line and water samples retrieved from the dump tank before and after the LOCA tests provide information on the extent of the release from the pressure flask in the test rig. The results of tests 9, 10 and 11 all indicate that less than 1 % of the inventory of  $^{131}\text{I}$  and  $^{137}\text{Cs}$  is released from the flask, which in comparison with the NRC guide point to low release for the fragmented high burnup fuel employed in the experiments. See table 1. The release in test nr.9 is somewhat higher than the later tests, which is no surprise considering the high burnup and the fuel relocation in that test.

Table 1: Fraction of estimated inventory released from the test rig.

Nuclide	Test nr.		
	Nr.9	Nr.10	Nr.11
$^{131}\text{I}$	0.4 %	0.02 %	0.06 %
$^{137}\text{Cs}$	0.1 %	0.06 %	0.04 %

### 3.4 GAMMA SCANNING MEASUREMENT SET-UP

The gamma scanning technique for determination of activity inventory in a fuel rod relies on high resolution gamma-ray spectroscopy measurements on the fuel stack of a rod and correlation of the measured activities to a calibrated standard rod. In the HBWR laboratories the rod or pressure flask subject to examination is mounted vertically in a rig installed in a hot cell. The rig can elevate the rod and move it laterally. The measurement equipment consists of collimators installed in a built-in penetration to the hot-cell and a High Purity Germanium (HPGe) detector with accompanying electronics and spectroscopy software. The distance from the fuel rod to the detector can be adjusted to suit the gamma intensity from the fuel rod and the selected collimation.

### 3.5 THE MEASUREMENT TECHNIQUE AND THE RELEVANT GAMMA RADIATION

In the case of inventory determination of a LOCA test fuel rod, the collimator geometry is such that an axial section of the fuel stack of 0.4 mm is exposed to the detector. Gamma-ray spectra are collected at several positions over the full axial length of the rod, typically 10 to 50 data points. The centre of attention is the 637.0 keV gamma peak of  $^{131}\text{I}$  and the 661.7 keV gamma peak of  $^{137}\text{Cs}$ , see table 2 for information concerning the relevant nuclides and gamma peaks. As a consequence of the short 8 day half-life of  $^{131}\text{I}$ , the scan is usually completed within 14 days of reactor shutdown. The LOCA fuel in the scan rig is then replaced by a standard rod, with known activity amount and distribution of  $^{137}\text{Cs}$ . A counting efficiency factor of the measurement system can then be determined for the 661.7 keV gamma peak of  $^{137}\text{Cs}$ . In the next step, the standard rod is replaced by a second reference rod, typically a driver fuel rod irradiated to a burnup of 30-40 MWd/kgU. In such a rod  $^{134}\text{Cs}$  will be a major constituent of the radioactive inventory. The  $^{134}\text{Cs}$  nuclide is the source of several gamma peaks, most importantly 604.7 keV and 795.8 keV. As the counting efficiency factor of the measurement system is almost linear with energy in the range from 600 keV to 800 keV (Fure, 2007), and the counting efficiency factor at 661.7 keV has already been determined, the counting efficiency factor at 637.0 keV of  $^{131}\text{I}$  can be determined by making use of the major  $^{134}\text{Cs}$  peaks. Finally the various counting efficiency factors found can be applied to the LOCA rod data set, and the inventory of  $^{131}\text{I}$  and  $^{137}\text{Cs}$  determined.

Table 2: Table lists relevant gamma peaks. Source is the Nucléide-LARA web database by Laboratoire National Henri Becquerel (LNHB).

Energy (keV)	Emission Prob. (%)	Nuclide	Half-life
604.7	97.6	$^{134}\text{Cs}$	2.07 y
637.0	7.26	$^{131}\text{I}$	8.02 d
661.7	85.0	$^{137}\text{Cs}$	30.1 y
795.8	85.5	$^{134}\text{Cs}$	2.07 y

The 637.0 keV peak of  $^{131}\text{I}$  is partly overlapping with the 635.9 keV peak of  $^{125}\text{Sb}$  and the 636.2 keV peak of  $^{151}\text{Pm}$ . However, in the case of the LOCA tests the total contribution to the  $^{131}\text{I}$  peak has been estimated, using Origen software, to cause an insignificant systematic error well below 1 %. The 661.7 keV peak of  $^{137}\text{Cs}$  poses few problems in the analysis of the LOCA test fuel data. If the amount of  $^{131}\text{I}$  is substantial, its 667.7 keV peak may interfere with the  $^{137}\text{Cs}$  peak. However, this analysis challenge can be managed by some days of decay before the measurements are initiated. The 657.8 keV peak of  $^{110\text{m}}\text{Ag}$  is also clearly present both in the LOCA fuel and in the standard reference rod, but will not overlap significantly with the  $^{137}\text{Cs}$  peak. Yet the  $^{110\text{m}}\text{Ag}$  peak must be handled with care when analysing the data. The driver fuel rod has typically been in decay for half a year or so before use as a  $^{134}\text{Cs}$  reference rod. Several nuclides are the source of gamma peaks in the range from 600 to 610 keV. However, in addition to the 604.7 keV gamma peak of  $^{134}\text{Cs}$ , the only two noteworthy peaks after some decay time have  $^{125}\text{Sb}$

as the source nuclide. Both peaks are relatively small, but must be considered with care when evaluating the 604.7 keV data of the driver rod. Finally, there are no significant gamma peaks interfering with the analysis of the 795.8 keV peak of  $^{134}\text{Cs}$ .

### 3.6 POST-LOCA $^{137}\text{Cs}$ AND $^{131}\text{I}$ IN ROD FUEL

Determination of the activity of  $^{131}\text{I}$  and  $^{137}\text{Cs}$  by gamma scanning technique necessitates fuel geometry similar to the calibration references. The limited fuel relocation in test rods 10 and 11 were acceptable for the application of the gamma scanning technique: All data points in both tests were included in the data sets to be analysed. In the case of test nr.9 the geometry requirement was approximately satisfied for only 12 out of 24 data points. However, the 12 data points were not evenly distributed over the active fuel length. As the LOCA experiment power profile is peaked and slightly asymmetric, an additional uncertainty is thus introduced when the selected data points are extrapolated to represent the total fuel amount. Notwithstanding these complications and uncertainties, an analysis of the activity inventory in the more intact parts of the fuel stack was tried. Due to the extensive fuel relocation, involving the entire fuel stack except for the top pellets, a supplementary approach based on test nr.8 was employed in the  $^{131}\text{I}$  and  $^{137}\text{Cs}$  activity evaluation of test nr.9. Gamma scan data of test nr.8 provided nuclide specific ratios describing the total activity of the fuel relative to the activity in the top end pellets only (Eitrheim, 2010). As the power profiles of test nr.8 and test nr.9 were quite similar, it is assumed that the nuclide specific ratios of test nr.8 were valid also in the case of test nr.9. Based on the activities measured in the top end pellets, the  $^{131}\text{I}$  and  $^{137}\text{Cs}$  inventory of the entire fuel in test nr.9 were found. Fractions of  $^{131}\text{I}$  and  $^{137}\text{Cs}$  in the fuel rod relative to the estimated total are summarised for the three relevant tests in table 3. Although these results are affected by uncertainties as discussed above, they point to low  $^{131}\text{I}$  release for the fragmented high burn up fuel employed in the experiments. For test nr.11 the measured activity inventory in the fuel rod is greater than the Origen estimate; however the results are within the uncertainty limits. Combined, the results of tables 1 and 3 indicate that most of the activity released from a rod remains in the pressure flask.

Table 3: Fraction of  $^{131}\text{I}$  and  $^{137}\text{Cs}$  remaining in the fuel rod post-LOCA, determined using Origen software and gamma scanning technique.

Nuclide	Test nr.		
	Nr.9	Nr.10	Nr.11
$^{131}\text{I}$	98 % and 86* %	$96.4 \pm 7$ %	$105 \pm 7$ %
$^{137}\text{Cs}$	93 % and 81* %	$94.6 \pm 6$ %	$105 \pm 6$ %

\*Results based on the nuclide specific ratios found in test nr.8

### CONCLUSION

So far, the relevant Halden LOCA tests indicate that less than 1 % of both  $^{131}\text{I}$  and  $^{137}\text{Cs}$  is released from the test rig. The analysis of the  $^{131}\text{I}$  and  $^{137}\text{Cs}$  in the fuel itself has proved more difficult, among other factors because of uncertainties related to the damaged rod geometry. Although these results are affected by uncertainties as discussed, they do point to low iodine release for the fragmented high burnup fuel employed in the experiments. With some refinement the gamma scanning technique for determination of the  $^{131}\text{I}$  and  $^{137}\text{Cs}$  inventory seems suitable, especially in cases with limited fuel relocation.

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