Assessment of Radioactivity Inventory in Swedish LWRs at Time of Decommissioning – 14279

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ABSTRACT

Decommissioning studies for nuclear power reactors are to be performed in order to assess the decommissioning costs and the waste volumes, as well as to provide data for the licensing and construction of the LILW repositories. An important part of this work is to estimate the amount of radioactivity in the different types of decommissioning waste.

Studsvik has performed these assessments for all Swedish NPPs as well as other nuclear facilities in Sweden using thorough on-site sampling and robust calculations developed by Studsvik's team of senior experts. Precision has been found to be relatively high close to the reactor cores, but then declines as distance from the core increases.

The decommissioning waste from a LWR can be separated into different categories such as:

- Material affected by the neutron flux from the reactor core
- Process systems
- Waste handling systems
- Contaminated structures

The determined specific activities for different systems (or part of systems) are combined with data on weights and contaminated surface areas in order to assess the total activity.

A key issue in the assessments has been efforts to reduce the uncertainties. Combining the unique knowledge in assessment of radioactivity inventories, the large data bank the waste processing represents and the knowledge and records from the laboratories, the activity determination codes can be validated and the waste processing analysis supported with additional data.

INTRODUCTION

Studsvik has been responsible for the determination of activity inventories at decommissioning for all the Swedish nuclear power reactors, in total 13, of which three are permanently shut down and ten operating.

Reactors permanently shut down:

NPP Barsebäck 1 and 2 (BWR, closed 1999/2005)
Project "RivAkt", Total activity assessment (2007), evaluation of performed system decontaminations (2008), improved assessment of activity in reactor internals (2012)

Ågesta (PHWR, closed in 1974)
Total activity assessment (2010)

Reactors in operation:

NPP Ringhals 1, 2, 3 and 4 (BWR + 3x PWR)
Total activity assessment (2007), updates (2010, 2012)

NPP Forsmark 1, 2 and 3 (BWR)
Total activity assessment (2010), update (2012)

NPP Oskarshamn 1, 2 and 3 (BWR)
Total activity assessment (2010), update (2012)

RADIOACTIVITY INVENTORY ASSESSMENTS

Prerequisites

The prerequisites for the performed radioactivity inventory assessments are summarized in the following way:

- The total operation time for each reactor is based on plant specifications. This means actual operation times for the Barsebäck and Ågesta reactors, and predicted times, 40, 50 or 60 years, for the reactors still in operation.
- A decay period of at least one year is presumed. It means that nuclides with half-lives shorter than one year have been disregarded.
- Operational waste such as spent fuel, ion exchange resins and filter media is assumed to have been removed prior to the decommissioning. However, small amounts of waste are assumed to remain in the waste handling systems. No major decontamination campaigns are considered prior to decommissioning, except for the Barsebäck plants where performed decontaminations campaigns are considered.
- Only plant materials with activity contents expected to exceed the exemption levels are included in the assessment.

Input to Activity Assessment

The main inputs to the activity assessments are:

- Safety Analysis Report (SAR) data describing activity inventories in the plants. Many of these SAR reports have recently been updated in connection to power uprate and modernization projects, i.e. are reflecting most actual operating conditions for the plants. Measured plant data such as dose rate and gamma scan measurements during outage conditions, reactor water data, moisture content in steam (for BWRs), and data describing the fuel leakage history.
- Components weights and surface areas in contact with active process media. These weights and areas are broken down into system “idents” describing system or part of system with certain activity conditions. Future operation conditions such as total operation time, planned modifications (e.g. power uprates), reference time for decommissioning, etc.

Source Terms Considered

The following types of source terms are considered in the assessments:

- Neutron induced activity in reactor internals, reactor pressure vessel (RPV), RPV insulation and biological shield of concrete and reinforcement surrounding the RPV. Neutron fluxes in the components are determined by calculations with the 3D neutron transport code MCNP. Compositions of the different materials, including trace elements such as Co, are based on materials specifications, materials certificates, and general information about materials compositions. Neutron fluxes, materials compositions, neutron activation cross sections and operation history are combined to calculated activity inventories with the use of different computer models (IndAct, FISPACT).
- Activated corrosion products on system surfaces, so called “crud”. The determination of contamination level on surfaces in the primary circuit, i.e. surfaces in contact with hot reactor water, is based on developed calculation models, CrudAct, for BWRs and PWRs, which are well benchmarked against measured reactor data. The resulting nuclide vectors are distributed between different reactor system idents in relation to measured relative contamination levels of different parts of systems, the primary circuit acting as reference.
- Fission products and actinides from leaking fuel. SAR leakage models are combined with measured activity data from the plants. Of special interest are the cases with fuel dissolution from rather open fuel failures, where the reactor coolant is in direct contact with the fuel material. Such fuel release turns out to have a significant memory effect in form of uranium contamination on core (so called tramp U), and actinide incorporation in the oxide layers formed on system surfaces. The tramp U causes production of short-lived noble gases that results in noble gas daughter accumulation, Sr-90, Cs-135, Cs-137, etc., in the off-gas decay systems.
- System leakage results in some accumulation of activity in affected building areas of concrete in the plant. This contamination reflects the nuclide composition in the reactor coolant.

Nuclides Considered

A review was initially performed, identifying the following 28 nuclides that were covered in the reported decommissioning inventories: H-3, C-14, Cl-36, Ca-41, Fe-55, Ni-59, Co-60, Ni-63, Sr-90, Nb-94, Tc-99, Ag-108m, Cd-113m, Sb-125, I-129, Cs-134, Cs-135, Cs-137, Sm-151, Eu-152, Eu-154, Eu-155, Pu-238, Pu-239, Pu-240, Pu-241, Am-241, and Cm-244.It has later been identified a need to add some extra nuclides in order to coordinate the nuclide list for decommissioning waste with the list used for operational waste. For that reason another 20 nuclides are included in the 2012 update for the Swedish plants. They are: Be-10, Se-79, Mo-93, Zr-93, Nb-93m, Ru-106, Pd-107, Sn-126, Ba-133, Pm-147, Ho-166m, U-232, U-236, Np-237, Pu-242, Am-242m, Am-243, Cm-243, Cm-245, and Cm-246.

Some other nuclides have also been discussed, e.g. the nuclide Fe-60 (half-life, $T_{½}$ = 1.5 million years, My), which is mainly produced via neutron reactions in iron.

The daughter nuclide Co-60 contributes to a high dose factor for Fe-60 both for ingestion and inhalation. This is illustrated in the Table I below where the specific activity in typical BWR crud has been calculated with the computer code FISPACT-2007. A decay period of 1000 years has been considered, and the activity has been recalculated to ingestion and inhalation doses per gram fuel crud. The nuclide Fe-60 is far from dominating, but ends up on the Top10 list. It shall be noted that different nuclides transport properties in the environment is not considered in the assessment, which of course may change different nuclides importance considerably.

An overall conclusion is that the introduction of additional nuclides will imply a need for improved evaluation and validation processes.

TABLE I Calculated radio toxicity in typical BWR fuel crud after decay 1000 years (FISPACT-2007).

Nuclide	**Ingestion**	**Nuclide**	**Inhalation**
	mSv/g		mSv/g
Ni-59	1.3E-01	Nb-94	3.4E+00
Nb-94	1.2E-01	Ni-59	9.0E-01
Ni-63	4.2E-02	Ni-63	3.6E-01
Mo-93	1.9E-02	Zr-93	1.8E-02
Zr-93	7.8E-04	Mo-93	1.4E-02
Nb-93m	7.2E-04	Nb-93m	1.1E-02
Tc-99	2.5E-04	Tc-99	5.2E-03
Fe-60	1.4E-04	Fe-60	3.7E-04
Re-186m	6.8E-06	Nb-91	7.1E-05
Total	3.1E-01	Total	4.7E+00

VALIDATION OF THE INVENTORY ASSESSMENT MODELS

Below are presented some examples of validation projects that have been or are planned to be used in the assessments of decommissioning activity inventories.

Ringhals-3 Steam Generator

Removed steam generators (SGs) from the Ringhals PWRs have been transported to Studsvik for treatment. The resulting activities from one of the R3 SGs in produced waste and ingots are summarized in Table II. Some special measurements were made in Studsvik, e.g. for the nuclide Ni-63. Three samples of the waste from the blasting were sent to the University of Lund for analysis of C-14 with the same method that is used for determination of C-14 in ion exchange resins.

The Co-60 activity in the waste is compared to the activity inventory estimated from in-plant gamma scans. It is noted that the waste activity is higher than the activity based on the gamma scans. This difference is likely due to the complicated geometry for the evaluation of the gamma scans, especially when measuring activity in the Inconel tubes from the outside of the SG.

The measured Ni-63/Co-60 ratio, about 0.1, is in line with earlier assessments. The detected C-14 activity is, however, not considered in the earlier assessments. The detected amount of C-14 corresponds to about 0.24 % of the production in the reactor coolant during a year.

TABLE II R3 SG – Measured activity from treatment in Studsvik compared to Co-60 activity inventory based on in-plant gamma scanning.

Measured activity after processing in Studsvik, Bq				
Nuclide	**Melted metal**	**Blasting residues**	**Other waste**	**Total**
Co-60	2.2E+11	2.3E+12	1.1E+12	3.6E+12
Ni-63	2.2E+10	2.2E+11	1.0E+11	3.5E+11
Ni-63/Co-60				0,098
C-14	5.2E+07	5.7E+08	2.8E+08	9.0E+08
C-14/Co-60				0.00025

Reference date: 1995-06-01

In-plant gamma scanning results, Bq			
Nuclide	**Inconel 600**	**Stainless steel**	**Total**
Co-60	8.8E+11	9.2E+10	9.7E+11

Reference date: 1995-06-01

Barsebäck 1 and 2 Decontamination Campaign

An example of important field data that have been used in the decommissioning studies is shown below in Table III. Three large decontamination campaigns of the primary circuit have been performed in Barsebäck BWRs (B1 and B2), and the removed activity has been carefully recorded. Hard-to-measure nuclides such as Ni-59 have been determined. This is valuable for validation of the calculation models. Furthermore, the measurement of actinides removed from the system surfaces can be correlated to the plants fuel failure history. B1 has been practically free from failures, while B2 had a fuel failure in 1992 resulting in a release of about 10 g of uranium. Actinides corresponding to about 1 g of uranium are found on system surfaces after about 10 years of operation, i.e. the incorporation of actinides in the system oxides has a long memory effect that has to be considered.

TABLE III. Measured activity removed in three decontamination campaigns in B1 and B2.

	B1/2008 [Bq]	B2/2007 [Bq]	B2/2002 [Bq]	Total [Bq]
Co-60	1.33E+12	2.13E+12	7.55E+11	4.21E+12
Fe-55	6.72E+11	1.28E+12	6.69E+11	2.42E+12
Mn-54	8.01E+08	3.98E+10	7.91E+08	4.14E+10
Ni-59	1.68E+09	1.18E+09	1+3E+09	4.50E+09
Ni-63	2.13E+11	1.59E+11	2.13E+11	5.86E+11
Sb-125	2.30E+10	6.60E+10	2.44E+10	1+3E+11
Tc-99	8.44E+05	3.25E+05	4.48E+05	1.62E+06
Pu-238	3.41E+06	4.69E+06	1.52E+07	2.33E+07
Pu-239	4.13E+05	5.44E+05	1.76E+06	2.72E+06
Pu-240	6.75E+05	8.89E+05	2.87E+06*	4.44E+06
Pu-241	1.07E+08	1.83E+08	5.93E+08	8.83E+08
Am-241	1.57E+06	4.03E+05	1.30E+06	3.28E+06
Cm-244	3.56E+06	5.79E+06	1.87E+07	2.81E+07

* Memory effect of fuel dissolution in B2 in 1992 (totally approximately 10 g U).

Barsebäck 1 – Activity in Bioshield and RPV Insulation

Another example of a performed validation is the sampling and activity measurements on the biological shield and RPV insulation performed in the B1 plant, which is compared to measured data in the below Table IV. The calculated values show slightly higher level than calculated, i.e. a certain degree of conservatism is maintained in the earlier assessment. These measurements are used in the validation of ongoing refined modeling of the neutron activation in reactor internals, pressure vessel and bioshield.

TABLE IV B1 – Comparison between measured and calculated activity in RPV insulation and biological shield.

	Caposil [Bq/kg]		**Aluminum Sheet** [Bq/kg]	
Nuclide	**Calculated**	**Measured**	**Calculated**	**Measured**
Co-60	3.3E+05	2.4E+05	8.4E+04	6.3E+04
Cs-134	1.4E+05	4.2E+04		
Mn-54	5.6E+05	5.2E+05	3.2E+04	2.0E-4
Zn-65			1-6E+05	6-3E+04
	Concrete [Bq/kg]		**Reinforcement** [Bq/kg]	
Nuclide	**Calculated**	**Measured**	**Calculated**	**Measured**
Co-60	7.6E+05	3.0E+05	2.7E+07	6.2E+06
Mn-54			1.3E+07	5.3E+06
Cs-134	9.0E+04	5.5E+04		
Eu-152	1.8E+06	1.3E+06		
Eu-154	1.6E+05	1.2E+05		

Forsmark BWRs –Turbine Components Being Treated in Studsvik
Totally about 1300 tons of turbine components were removed from the Forsmark BWRs (F1, F2 and F3) during the period 2004–2006 and sent to Studsvik for treatment. A summary of the weights and Co-60 and Cs-137 activities in different categories is shown in Table V. About 95 % of the material was subject to clearance, 71 % for direct clearance and the remaining 24 % after some additional decay. The remaining 5 %, process waste, is to be disposed.

The total Co-60 activity, 1.6 GBq, is in line with performed assessments based on plant data. However, the measured Cs-137 activity, 0.07 GBq, is higher than performed assessments. The source of Cs-137 in this case is expected to be from decay of short-lived Xe-137 ($T_{½}$ = 3.8 minutes) which is predominantly from tramp uranium on the core. Cs-137 activity from decay of Xe-137 is considered in the off-gas systems, but has not earlier been considered on the turbine components.

TABLE V F1, F2 and F3 – Turbine components - Weights and activity in different categories after treatment in Studsvik.

	Weight [Mg]	**Co-60** [Bq]	**Cs-137** [Bq]	**Specific activity** [Bq/g]
Direct clearance[1]	940	4.3E+08		0.46
Clearance after decay period[2]	316	8.6E+08		2.7
Waste[3]	64	3.0E+08	9.3E+07	6.1
Total	1320	1.6E+09	9.3E+07	1.3

1 Direct clearance (<1 Bq/g)
2 Clearance after period of decay (until <1 Bq/g)
3 Process waste

CONCLUSIONS

A number of conclusions could be drawn based on the current situation.

There is a driving force for validation of the activity determination codes. An improved knowledge of the nuclide inventory in both ILW and LLW will reduce uncertainties. Reduced uncertainties may simplify the safety analysis reports as well as the qualification/ licensing and the design of the disposal sites.

Current status:

- Good calculation models exist.
- Validations of calculated activity assessments have been performed in certain areas, mainly for easy to measure nuclides – good results and deep understanding.
- The clearance route is in general well validated.
- The most important validations, in a repository long term perspective, are often weak if started up. Significant differences between countries.
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- Optimized screening, to sort out nuclides of no importance, is of significant value. Which nuclides that can be sorted out depend on the actual disposal and repository situation.