Validation of activity determination codes and nuclide vectors by using results from processing of retired components and operational waste

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Paper for Workshop on “Radiological characterisation for Decommissioning” Studsvik, Sweden 17-19 April 2012

Abstract

Decommissioning studies for nuclear power reactors are 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 ALARA Engineering has performed such assessments for LWRs and other nuclear facilities in Sweden. These assessments are to a large content depending on calculations, senior experience and sampling on the facilities. The precision in the calculations have been found to be relatively high close to the reactor core. Of natural reasons the precision will decline with the distance. Even if the activity values are lower the content of hard to measure nuclides can cause problems in the long term safety demonstration of LLW repositories.

At the same time Studsvik is processing significant volumes of metallic and combustible waste from power stations in operation and in decommissioning phase as well as from other nuclear facilities such as research and waste treatment facilities.

Combining the unique knowledge in assessment of radioactivity inventory and the large data bank the waste processing represents the activity determination codes can be validated and the waste processing analysis supported with additional data.

The intention with this presentation is to highlight how the European nuclear industry jointly could use the waste processing data for validation of activity determination codes.

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Introduction

Short about Studsvik

Studsvik, previously named AB Atomenergi, was founded in 1947 as a state own company with the mission to develop and operate nuclear power stations in Sweden. Today Studsvik is a commercial company focused on providing services to the international nuclear industry.

A key knowledge which has remained over the years in the company is the deep knowledge in characterisation of materials and radioactive waste (even though radioactive waste management not was in spot as much in the 1950s as it is today). The competence in radiological characterisation was further strengthened when ALARA Engineering AB joined the Studsvik Group.

Studsvik has built several theoretical and practical references in the area of radiological characterisation of materials, systems and buildings for decommissioning.

Furthermore Studsvik has processed radioactive waste for a few decades focusing on volume reduction of organic waste and aiming for clearance of metals after treatment.

Overview – initial facts

A known fact is that all radioactive objects need to be characterised and managed. Depending of the properties and the radioactive content the material can be subject to clearance directly or after treatment, put into a form suitable for disposal and disposed.

Critical parameters for characterisation differ between routes. This has to be considered already in the validation planning.

Quality assurance of activity assessment calculations and nuclide vectors requires validation require physical measurements.

Normally characterisation for clearance is easier than characterisation for disposal as hard to measure nuclides are of less importance for clearance.

For clearance based on European Commission recommendations RP 89, RP 113 and RP 122 part 1 several low emitting beta nuclides can be screen out as they have high clearance levels.

For waste to be disposed several nuclides which is of concern in terms of clearance can be screened out in the long term safety calculations as they will decay to almost nothing in a relatively short period of time. They may however be of concern from an operational safety point of view.

Good understanding, validated and documented nuclide vectors and assessment calculations supports graded approach – saves money and time

Well developed areas

- Activity assessment for easy to measure nuclides (gamma) inside and close to the reactor pressure vessel of NPPs.
- Characterisation of contaminated metals subject to free release (clearance) either directly or after treatment.
- Screening of nuclides in the clearance process i.e. nuclides without importance for clearance

Areas with development potentials

- Validation of calculation models for hard to measure nuclides.
- Validation of activity assessment calculations for systems with low contamination levels far from the reactor grid.
- Nuclide composition after treatment for in a repository perspective critical hard to measure nuclides (such as C-14 and Cl-36).
- Characterisation models for nuclear facilities other than NPPs and where nuclide vectors have varied by time.
- Nuclide screening models for waste to be disposed

Key parameters for validation

Essential for any type of validation program is good knowledge of the conditions and quality assurance. For validation of theoretical models by sampling and analysis either of the material to be processed or usage of the processing residues it is very important to secure the traceability and to define the comparability to avoid that incorrect conclusions are drawn.

It is of high importance with good understanding of process parameters such as:

- How the materials, systems and buildings have been contaminated
- Variations in operations and how it has affected the activity build up
- The way waste treatment affects the nuclide inventory and how treatment effects can simplify the validation process

The properties of the different nuclides require different approaches and methods to estimate the radioactivity in the physical material.

A few examples related to iron based metals:

- Co-60, Fe-55, Ni-59/63 and other activation products will even though just occurring as contamination to a large percentage alloy with the steel. This means that these nuclides will be present in decontamination waste, in the metal ingots as well as in the slag and dust from the melting process. This is no problem for gamma-emitting nuclides as Co-60 but makes the activity determination a lot more complex for the low energy beta emitters.

- Some volatile nuclides with a fairly high boiling point such as Cs-134/137 will boil off during melting but will be captured to almost 100% in the slag cover on the melting bath (if any) and in the dust filters. This means that Cs will be present in all but the metal ingots. The activity will then be significantly concentrated in the melting residues which is good for determination of the inventory.

- Heavy alpha emitters such as the uranium isotopes and the transuranium element will be transferred to the slag which means that they will be significantly concentrated in the slag i.e. makes the determination of the activity easier.

- C-14 and to certain extent H-3 and the Iodine isotopes are a lot more complex to estimate in the thermal processing stage. Most likely these nuclides should be determined by sampling prior to melting. Either by sampling prior to start of treatment of by analysis of the decontamination residues.

Key issues for a successful validation

- Review documentation of the nuclide vector development including boundary conditions.
- Define objectives and purpose of validation.
- Analyse the distribution and behaviour of the different nuclides i.e. identify uncertainties in using nuclide vectors. Make sure all process parameters are understood.
- Identify and focus on key nuclides – perform a careful screening.
- Secure comparability and traceability - careful planning and selection of validation objects.
- Form and evaluate different fail test methods as tools for validation.

Determination of activity inventories

Studsvik ALARA Engineering has been responsible for the determination of activity inventories at decommissioning for most Scandinavian nuclear reactors. The reference list covers totally 15 reactors:

- Olkiluoto 1 and 2 (BWR)
  Process systems, database modification and update (2008)

- Barsebäck 1 and 2 (BWR)
  Project “RivAkt”, Total activity assessment (2007), evaluation of performed system decontaminations (2008), improved assessment of activity in reactor internals (2012)

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

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

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

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

Updates are on-going for most reactors where some additional radionuclides will be included.

Prerequisites

The prerequisites for the performed studies 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 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, 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. These assessments of component data have been performed by other organizations.

- 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 delay 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 28 nuclides that were covered in the reported decommissioning inventories:

- H3, C14, Cl36, Ca41, Fe55, Ni59, Co60, Ni63, Sr90, Nb94, Tc99, Ag108m, Cd113m, Sb125, I129, Cs134, Cs135, Cs137, Sm151, Eu152, Eu154, Eu155, Pu238, Pu239, Pu240, Pu241, Am241, Cm244

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 ongoing 2012 update for the Swedish plants:

- Be10, Se79, Mo93, Zr93, Nb93m, Ru106, Pd107, Sn126, Ba133, Pm147, Ho166m, U232, U236, Np237, Pu242, Am242m, Am243, Cm243, Cm245, Cm246

Some other nuclides have also been discussed, e.g. the nuclide Fe60 ($T_{½}$ = 1.5 My), which is mainly produced via neutron reactions in iron, mainly:

- Fe58 (n,γ) Fe59 (n,γ) **Fe60** (β^-) Co60m (IT) **Co60**

The daughter nuclide Co60 contributes to a high dose factor for Fe60 both for ingestion and inhalation. This is illustrated in the below table 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 Fe60 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.

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

Decay: 1000 y			
Nuclide	Ingestion mSv/g	Nuclide	Inhalation 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	Ag108m	3.3E-02
Ag108m	2.0E-03	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
Re186m	6.8E-06	Nb 91	7.1E-05
Total	3.1E-01	Total	4.7E+00

Examples of validation projects

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 the below table. Some special measurements were made in Studsvik, e.g. for the nuclide Ni63. Three samples of the waste from the blasting were sent to the University of Lund for analysis of C14 with the same method that is used for determination of C14 in ion exchange resins.

The Co60 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 Ni63/Co60 ratio, about 0.1, is in line with earlier assessments. The detected C14 activity is, however, not considered in the earlier assessments. The detected amount of C14 corresponds to about 0.24% of the production in the reactor coolant during a year. These measurements are presently being further evaluated.

R3 SG – Waste activity from treatment in Studsvik compared to Co60 activity inventory based on in-plant gamma scanning

Waste activity from processing in Studsvik

	Bq, 1995-06-01			
Nuclide	**Blasting**	**Melt**	**Other**	**Total**
Co60	2.27E+12	2.20E+11	1.07E+12	3.56E+12
Ni63	2.23E+11	2.16E+10	1.05E+11	3.50E+11
Ni63/Co60				9.83E-02
C14	5.68E+08	5.51E+07	2.77E+08	9.00E+08
C14/Co60				2.53E-04

In-plant gamma scanning on SS and Inc

	Bq, 1995-06-01			
Nuclide		**Inc600**	**SS**	**Total**
Co60		8.83E+11	9.20E+10	9.75E+11

Barsebäck 1 and 2 decontamination campaign

An example of important field data that have been used in the decommissioning studies is shown in the below table. 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 Ni59 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.

Measured activity removed in three decontamination campaigns in B1 and B2

ref.date	2007-11-01	2007-11-01	2007-11-01	2007-11-01
	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	1.63E+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	1.13E+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 about 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. The calculated values show slightly higher level than calculated, i.e. a certain degree of conservatism is maintained in the earlier assessment. These measurements will be used in the validation of ongoing refined modeling of the neutron activation in reactor internals, pressure vessel and bioshield.

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

	Caposil [Bq/kg]		Al sheet [Bq/kg]	
Nuclide	Calculated	Measured	Calculated	Measured
Co-60	3.3E5	2.4E5	8.4E4	6.3E4
Cs-134	1.4E5	4.2E4		
Mn-54	5.6E5	5.2E5	3.2E4	2.0E4
Zn-65			1.6E5	6.3E4

	Concrete [Bq/kg]		Reinforcement [Bq/kg]	
Nuclide	Calculated	Measured	Calculated	Measured
Co-60	7.6E5	3.0E5	2.7E7	6.2E6
Mn-54			1.3E7	5.3E6
Cs-134	9.0E4	5.5E4		
Eu-152	1.8E6	1.3E6		
Eu-154	1.6E5	1.2E5		

Forsmark BWRs –Turbine components being treated in Studsvik

Totally about 1300 tonnes 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 Co60 and Cs137 activities in different categories is shown in the below table. 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 Co60 activity, 1.6 GBq, is in line with performed assessments based on plant data. However, the measured Cs137 activity, 0.07 GBq, is higher than performed assessments. The source of Cs137 in this case is expected to be from decay of short-lived Xe137 ($T_{½}$ = 3.8 minutes) which is predominantly from tramp uranium on the core. Cs137 activity from decay of Xe137 is considered in the off-gas systems, but has not earlier been considered on the turbine components.

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

		Activity, Bq		Bq/kg
	Weight, kg	Co60	Cs137	Co60
Exempted[1]	940234	4.3E+08		4.6E+02
Ingots[2]	315745	8.6E+08		2.7E+03
Waste[3]	64290	3.0E+08	9.3E+07	4.7E+03
Total	1320269	1.6E+09	9.3E+07	1.2E+03

1 - Exempted ingots (limit Co60 <1000 Bq/kg)
2 - Ingots to be exempted after some decay
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 exists

- 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.

- Optimised screening, to sort out nuclides of no importance, is of significant value. Which nuclides that can be sorted out depends on the actual disposal and repository situation.

A way forward

Validation of activity determination codes could be costly especially if a project should cover a wide range of material from reactor interns to LLW far from the reactor core.

Validations can be of value for more than the actual facility as the target is to validate the codes. This means that other organisations than the specific facility owner may be interested in the results from a validation program.

Of these reasons we do propose that the following proposal is evaluated:

Form an international project for building up industry knowledge regarding activity assessment in metals from decommissioning to support validation and optimisation of radioactivity assessment codes.

Such a projects could be built on the principles of :

- Open books inside project
- joint financing
- common learning

Most likely there are material samples, used retired components sent for treatment and operational waste available that can be used as objects for the validations in combination with sampling and measurement in-situ.

Special areas of interest could be

- long lived beta-emitting nuclides
- nuclide distribution, behavior and effects of treatment
- optimisation of sampling and evaluation