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Title: "Assessment of activity inventories in Swedish LWRs at time of decommissioning"

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Decommissioning studies for the Swedish nuclear reactors are ongoing in order to assess the waste volumes and costs. Part of this work is assessing the amount of radioactivity in the various types of decommissioning wastes. ALARA Engineering has performed such assessments for most of the Swedish LWRs. The paper describes the methods used in these assessments and some of the main results.

Decommissioning studies

Decommissioning studies with the objective to assess activity inventories at the time of decommissioning have been performed by ALARA Engineering for the following Nordic nuclear reactors:

- Olkiluoto 1 and 2 (OL1/2): The assessment has covered activity in the process systems and updates have regularly been performed (the latest in 2008). In parallel, a modification and an update of a database describing the decommissioning work associated with the process systems have been performed.

- Barsebäck 1 and 2 (B1/2): A feasibility study named "RivAkt" was performed in 2002. In 2007 a total assessment of activity inventories was performed. Complete decontamination campaigns were performed in B2 and B1 in 2007 and 2008, respectively, and activity removed in the decontamination campaigns was evaluated in 2008.

- Ringhals 1, 2, 3 and 4 (R1/2/3/4). Full assessments of activity inventories were performed for these reactors (1 BWR, 3 PWRs) in 2007. An update considering changed total operation times and plant modernization programs is currently ongoing.

- Forsmark 1, 2 and 3 (F1/2/3): Full assessments of activity inventories were performed in 2010.

- Oskarshamn 1, 2 and 3 (O1/2/3): Full assessments of activity inventories were performed in 2010. An update of an earlier assessment for O3 considering the power uprate project PULS is under discussion.

- Ågesta: This pressurized heavy water reactor (PHWR) was permanently shut-down in 1974. An assessment of activity inventories is ongoing based on earlier studies in combination with the recent methods developed for other reactors.

Methods

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 B1/2 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 significantly 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.
- 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 offgas 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 of nuclides of potential interest for the decommissioning was carried out in the B1/2 RivAct project in 2002. Originally 65 nuclides were identified, that later on has been reduced to 30 nuclides as shown in **Table 1**. The table also indicates the main presence of the different nuclides.

***Table 1**: Nuclides considered in the decommissioning projects*

Nuclide	$T_{½}$	Main occurrence
H-3	12.33 y	Control rods (BWR), concrete
C-14	5.73 ky	Internals, waste
Cl-36	302.01 ky	Internals, concrete, waste
Ca-41	103.00 ky	Insulation, concrete
Mn54	312.5 d	Internals, RPV, crud
Fe-55	2.70 y	Internals, RPV, crud
Co-60	5.27 y	Internals, crud
Ni-59	74.95 ky	Internals, crud
Ni-63	100.04 y	Internals, crud
Sr-90	29.12 y	Waste, offgas systems
Nb-94	20.30 ky	Internals, RPV
Tc-99	212.86 ky	Internals, RPV, waste
Ag108m	418.00 y	Control rods (PWR), crud
Ag110m	249.9 d	Control rods (PWR), crud
Cd113m	14.09 y	Control rods (PWR)
Sb-125	2.73 y	Crud
I-129	15.69 My	Waste
Cs-134	2.06 y	Waste, concrete, insulation
Cs-135	2.30 My	Waste, offgas systems
Cs-137	30.00 y	Waste, offgas systems
Sm-151	88.73 y	Concrete
Eu-152	13.32 y	Concrete
Eu-154	8.60 y	Concrete
Eu-155	4.96 y	Concrete
Pu-238	87.70 y	Waste, crud
Pu-239	24.11 ky	Waste, crud
Pu-240	6.56 ky	Waste, crud
Pu-241	14.40 y	Waste, crud
Am-241	432.71 y	Waste, crud
Cm-244	18.10 y	Waste, crud

System "idents"

Based on nuclide vectors developed, activity inventories for the different system idents are calculated. An example of calculated data for three idents in the study for O2 is shown in **Table 2**. Some explanation to the different data:

- The three idents shown represent three different types:
  - 211.2 – RPV insulation – represents a case with only activity from neutron induced activity.
  - 321.3 – Part of system 321 – represents a case with only activity from surface contamination (crud).
  - 211.1 – RPV – represents a case with activity from both neutron induced activity and crud.
- Both total activity and surface activity (if any) are presented.
- An rough activity class is presented based on specific activity:
  - H - > 1 MBq/kg – Material with rather high radioactivity
  - L - >1 kBq/kg; < 1 MBq/kg – Material with rather low radioactivity, but likely above the exemption level
  - F - < 1 kBq/kg – The material may possibly be subject to exemption.

- A rough radiation level is calculated based on the specific activity for certain important nuclides:
  - Co-60, Mn-54, Cs-134, Cs-137, Eu-152

The total number of system idents is typically about 70.

***Table 2**: O2 – Three examples of system idents*
211.1 – RPV, 211.2 – RPV insulation, 321.3 – Part of system 321

Ident	**O2.211.1**		**O2.211.2**		**O2.321.3**	
Class [F/L/H]	**H**		**H**		**H**	
Rad [µSv/h]	3.2E+03		4.3E+01		6.5E+02	
Activity [Bq]	4.8E+13		8.5E+09		1.6E+11	
Activity [Bq/kg]	7.8E+07		1.7E+06		7.2E+06	
Unit	**[Bq/m^2]**	**[Bq]**	**[Bq/m^2]**	**[Bq]**	**[Bq/m^2]**	**[Bq]**
H-3				3.8E+05		
C-14	0	2.7E+09		1.9E+07		
Cl-36	0	2.1E+06		1.5E+07		
Ca-41				4.0E+08		
Mn54	4.9E+08	3.3E+12		2.9E+08	1.2E+07	4.2E+09
Fe-55	5.7E+09	3.4E+13		7.2E+09	1.4E+08	4.9E+10
Co-60	5.5E+09	5.5E+12		5.6E+08	1.4E+08	4.7E+10
Ni-59	5.1E+07	4.2E+10		2.4E+04	1.3E+06	4.4E+08
Ni-63	6.8E+09	5.1E+12		2.3E+06	1.7E+08	5.9E+10
Sr-90						
Nb-94	0	8.8E+07				
Tc-99	3.9E+03	5.1E+07			9.8E+01	3.4E+04
Sb-125	4.2E+07	2.4E+10			1.1E+06	3.6E+08
I-129						
Cs-134				1.0E+08		
Cs-135						
Cs-137						
Eu-152						
Eu-154						
Eu-155						
Pu-238	1.0E+05	5.7E+07			2.5E+03	8.8E+05
Pu-239	9.0E+03	5.1E+06			2.3E+02	7.8E+04
Pu-240	1.2E+04	6.5E+06			2.9E+02	1.0E+05
Pu-241	3.6E+06	2.1E+09			9.1E+04	3.1E+07
Am-241	5.2E+03	2.9E+06			1.3E+02	4.5E+04
Cm-244	1.6E+05	8.9E+07			3.9E+03	1.4E+06
	Source	**Factor**	**Source**	**Factor**	**Source**	**Factor**
Surface	Crud_321	2	0	0	Crud_321	0.05
Volume	I_211_1		I_211_2		0	
Area [m^2]	563		0		346	
Weight [kg]	620000		5000		22395	
[µSv/h per Bq/kg]	3.0E-04		3.0E-04		3.0E-04	
Nuclide	Co60		Co60		Co60	

Validation

As identified earlier the calculated activity inventories are based on a significant amount of field records. Some examples:

The fuel leakage models are based on benchmarking model data against measured data, an example shown in **Figure 1**. The same models are used for updated SARs, and the predicted leakage levels in the future operation are based on the realistic levels as specified in the SARs. Recent reactor operation is characterized by great caution with respect to operation with fuel failures, i.e. also the realistic SAR levels are likely somewhat conservative.

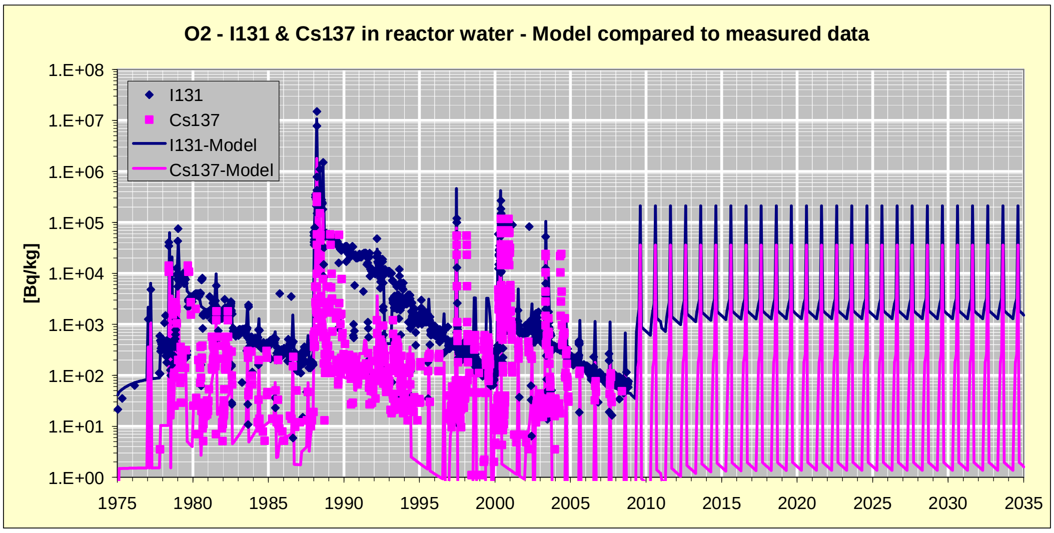

***Figure 1**: O2 – Measured reactor water concentrations of I-131 and Cs-137 as basis for the fuel leakage model*

An example of important field data that have been used in the decommissioning studies is shown in **Table 3**. Three large decontamination campaigns of the primary circuit have been performed in B1/2, 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. Furthemore, 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 about 5 g of tramp U on the core. 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 3**: Measured activity removed in three decontamination campaigns in B1/2*

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 (about 5 g of Tramp U

Another example 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 **Table 4**. The calculated values show slightly higher level than calculated, i.e. a certain degree of conservatism is maintained.

***Table 4**: 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		

Example of calculated inventories

A summary of calculated activity inventories in different parts of the O2 plant is shown in **Table 5** as an example. The total activity is about 38000 TBq, with a waste weight of about 3400 tonnes. The activity is dominated by the reactor internals, while waste volumes are rather equally distributed between reactor and turbine building. Dominating radionuclides are Fe-55, Co-60 and Ni-63, of which Co-60 by far dominates the radiation fields. Sr-90 and Cs-137 are important radionuclides in the offgas systems, and tritium and Eu isotopes in activated concrete in the biological shield.

The main activity is found in the reactor internals, where the uncertainty is estimated to be a factor of 2. The farther from the reactor, the lower the activity, but with increasing relative uncertainty, up to an order of magnitude. This uncertainty can have large impact on which system parts that could be exempted, i.e. the total waste volume. Note, however, no system decontaminations have been assumed, i.e. waste volumes are likely to be reduced through decontamination campaigns.

***Table 5**: O2 – Summary of calculated activity inventories (decay time 1 y)*

	Activity inventory [Bq]								
	Reactor building (R)					Turbine building (T)			O2
	Concrete	RPV Internals	Pool walls	System 3xx	Total	332, 341, 348, 552	4xx	Total	Grand total
H-3	4.7E+12	3.5E+14	0.0E+00	0.0E+00	**3.5E+14**	0.0E+00	0.0E+00	**0.0E+00**	**3.5E+14**
C-14	1.6E+09	4.6E+12	0.0E+00	1.5E+08	**4.6E+12**	1.3E+09	0.0E+00	**1.3E+09**	**4.6E+12**
Cl-36	4.9E+07	1.7E+09	0.0E+00	1.9E+05	**1.8E+09**	2.3E+04	0.0E+00	**2.3E+04**	**1.8E+09**
Ca-41	4.8E+09	4.0E+08	0.0E+00	0.0E+00	**5.2E+09**	0.0E+00	0.0E+00	**0.0E+00**	**5.2E+09**
Mn-54	4.6E+10	9.7E+14	1.4E+11	6.0E+11	**9.7E+14**	1.8E+09	5.7E+09	**7.5E+09**	**9.7E+14**
Fe-55	4.5E+12	2.9E+16	1.6E+12	3.5E+12	**2.9E+16**	6.0E+09	6.7E+10	**7.3E+10**	**2.9E+16**
Co-60	4.6E+11	4.3E+15	1.6E+12	7.5E+12	**4.3E+15**	2.3E+10	6.4E+10	**8.8E+10**	**4.3E+15**
Ni-59	1.2E+09	3.3E+13	1.5E+10	8.6E+10	**3.3E+13**	2.8E+08	6.1E+08	**8.9E+08**	**3.3E+13**
Ni-63	1.4E+11	4.0E+15	1.9E+12	1.0E+13	**4.0E+15**	3.2E+10	8.0E+10	**1.1E+11**	**4.0E+15**
Sr-90	4.7E+07	0.0E+00	0.0E+00	7.5E+09	**7.6E+09**	1.5E+10	0.0E+00	**1.5E+10**	**2.3E+10**
Nb-94	1.7E+07	5.0E+10	0.0E+00	4.7E+01	**5.0E+10**	7.4E+00	0.0E+00	**7.4E+00**	**5.0E+10**
Tc-99	7.4E+06	4.8E+10	1.1E+06	3.7E+08	**4.9E+10**	1.0E+07	4.6E+04	**1.1E+07**	**4.9E+10**
Sb-125	3.5E+08	1.0E+12	1.2E+10	3.8E+10	**1.1E+12**	7.2E+07	4.9E+08	**5.7E+08**	**1.1E+12**
I-129	3.3E+04	0.0E+00	0.0E+00	2.5E+06	**2.6E+06**	1.8E+06	0.0E+00	**1.8E+06**	**4.3E+06**
Cs-134	1.6E+10	1.0E+08	0.0E+00	9.9E+10	**1.1E+11**	1.4E+08	0.0E+00	**1.4E+08**	**1.2E+11**
Cs-135	1.4E+05	0.0E+00	0.0E+00	1.3E+07	**1.3E+07**	3.0E+06	0.0E+00	**3.0E+06**	**1.6E+07**
Cs-137	1.7E+10	0.0E+00	0.0E+00	1.3E+12	**1.3E+12**	6.0E+10	0.0E+00	**6.0E+10**	**1.4E+12**
Eu-152	3.3E+11	0.0E+00	0.0E+00	0.0E+00	**3.3E+11**	0.0E+00	0.0E+00	**0.0E+00**	**3.3E+11**
Eu-154	1.5E+10	0.0E+00	0.0E+00	2.1E+08	**1.5E+10**	2.8E+07	0.0E+00	**2.8E+07**	**1.5E+10**
Eu-155	6.7E+09	0.0E+00	0.0E+00	5.6E+07	**6.8E+09**	7.9E+06	0.0E+00	**7.9E+06**	**6.8E+09**
Pu-238	1.3E+05	4.1E+08	2.9E+07	2.8E+08	**7.2E+08**	4.3E+07	1.2E+06	**4.4E+07**	**7.6E+08**
Pu-239	3.5E+06	3.6E+07	2.6E+06	4.2E+07	**8.4E+07**	6.8E+06	1.1E+05	**6.9E+06**	**9.1E+07**
Pu-240	1.9E+04	4.6E+07	3.3E+06	5.3E+07	**1.0E+08**	8.6E+06	1.4E+05	**8.7E+06**	**1.1E+08**
Pu-241	1.9E+06	1.5E+10	1.0E+09	3.6E+09	**1.9E+10**	3.9E+08	4.3E+07	**4.4E+08**	**2.0E+10**
Am-241	8.0E+03	2.1E+07	1.5E+06	2.1E+07	**4.3E+07**	3.3E+06	6.1E+04	**3.4E+06**	**4.6E+07**
Cm-244	9.8E+04	6.3E+08	4.5E+07	1.8E+08	**8.6E+08**	2.2E+07	1.9E+06	**2.3E+07**	**8.8E+08**
Total	1.0E+13	3.8E+16	5.3E+12	2.3E+13	**3.8E+16**	1.4E+11	2.2E+11	**3.6E+11**	**3.8E+16**
Weight [kg]	**6.5E+05**	**7.8E+05**	**2.0E+05**	**5.4E+05**	2.2E+06	**3.8E+05**	**8.6E+05**	1.2E+06	3.4E+06