DANSK DEKOMMISSIONERING

Radiological Characterization and Decommissioning in Denmark

Danish Decommissioning (DD) is currently decommissioning the last Danish research reactor (DR3) and the Hot Cell facility.

The DR3 project will soon finish dismantling of the external parts of the reactor (January 2012). The approval for dismantling of neutron activated and tritium contaminated heavy water pumps and tubing was granted in December 2011.
DD will begin the work on the inner parts as the tendering process for equipment will start in 2012. Hereafter the dismantling of the top of the reactor will begin using the obtained remote controlled equipment.

The Hot Cell facility consists of 6 contaminated cells. The first cell have been opened and cleaned. Currently the work progresses by removing parts and hot spots from the other cells with the use of robotic equipment. Challenges, lack of conventional and radiological documentation, dose rates and contamination higher than expected and the confined space in the cells have delayed the project.

No final repository exists in Denmark. Therefore no official Waste Acceptance Criteria (WAC) have been formulated. However the Danish authority (SIS) does require a description of the waste in the interim storage facility (Inventory). Furthermore radiological characterisation of key nuclides is needed during decommissioning and dismantling. The information gained from the characterisation helps in the planning phase prior to the dismantling and for inventory calculations for later use. DD performs the radiological characterisation via both non-destructive and destructive analysis on samples.

The samples are measured with gamma spectroscopy using mathematical and geometrical analysis. Scaling factors are used for neutron activated waste (DR3) to determine the difficult-to-measure isotopes and pure beta emitters. The primary scaling isotope is Co-60. Waste from the Hot Cell facility is alfa contaminated and scaling procedures for determination of alfa contamination are currently used in the planning process. Scaling of alfa emitters will be incorporated into the inventory calculations.

Due to the variable nature of the systems being decommissioned, the sampling procedures are based on ad hoc principles. The number of samples needed is determined by the conventional characterisation of the systems. For systems where conventional knowledge is limited, more samples are generally needed earlier in the decommissioning process. Otherwise sampling can take place prior to the packing of the containers for the interim storage facility. In this case less sampling is needed as few representative samples for each material from each system in the container are sufficient.

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Introduction

In December 2000 the Danish reactor 3 (DR3) was shut down due to a suspected leak from the reactor tank. It was decided that the reactor would not be restarted and thus would have to be decommissioned. In March 2003 the Danish parliament decided that the decommissioning should begin. Danish Decommissioning (DD) was therefore founded with the purpose of decommissioning the 3 Danish research reactors (DR1, DR2, DR3), the Hot Cell facility and the waste treatment plant.

DD was divided from Risø National Laboratory (now DTU). The expertise was transferred from Risø to DD as DD's employees as the staff members were transferred. From the beginning it was necessary for DD to prove capable of decommissioning. The decommissioning of the first two reactors DR1 and DR2 was started shortly hereafter. The decommissioning of DR1 was done in 2004-2006 and of DR2 in 2006-2008. The experiences from these two projects are used in the DR3 and Hot Cell facility projects.

Figure 1, Overview image of the Risø peninsula and DD. The DR2 and the nuclear facilities DR3, Hot Cell and the Waste treatment plant are seen.

Hot Cell introduction

The decommissioning of the Hot Cell facility started in 2007 and is currently undergoing. The decommissioning is planned to be done in 2013, however the project have been delayed a year due to tendering problems for a remote controlled robot arm and is therefore not expected to be finished before 2014.

Figure 2, Preparation for removal of hot spots in the Hot Cell facility. The windows to the cells can be seen in the background.

The Hot Cell facility consists of 6 contaminated cells, where 2 of the cells are less contaminated compared to the others.
Historically the Hot Cell facility was used for both handling post irradiation experiments of fuel and source fabrication. When the facility was closed, the walls inside the cells were washed with water and the residue was collected in small piles. Over the years the hot spots in these piles irradiated the painting and in turn embedded themselves in the paint.

After the shut down the facility was embedded behind walls and an office space was built up around it while the knowledge of the facility was lost. The lead windows used during the operation of the facility was drained of oil and are not transparent anymore. Therefore information about the facility had to be collected by venturing inside the cells and from historical data.
The work has previously tended to be tediously slow due to the confined space, frequent high dose rates, recurring mock up sessions and lack of robotic equipment.
A 27 m long conveyor channel which connects the cells, are about to be dismantled. The conveyor will need to be cut up inside the first cell. The conveyor channel is expected to contain several different hot spots.

DR3 introduction

The internal parts of DR3 are to be decommisioned in 2012-2016. The last of the support systems on DR3 will be decommissioned at the first half of 2012. The dismantling of the so called heavy water room, consisting of pumps, valves and tubing, underneath the core of the reactor was approved by the authorities in December 2011. Detailed work plans for the dismantling work is currently being developed. For DR3 most of 2012 will consist of planning, designing of needed tools and tendering.

The tubes, valves and pumps were the primary heavy water circuit and are thus expected to be contaminated with tritium. As most of the waste will be steel, it is expected that the majority of the waste can be sent to an external contractor for meltning. The legal tendering implications for this are currently being investigated as this will be our first experience of sending waste to an external contractor. The heavy water room will be the last of the external systems on DR3.

Sampling or Modelling

One must determine if the standard of determining the activity in the nuclear facility or waste is based on sampling procedures or models or a combination of these. DD uses extensive sampling of each dismantled waste system.

No final repository exists in Denmark. Therefore no official Waste Acceptance Criteria (WAC) have been formulated. However the Danish authority (SIS) does require a description of the waste in the interim storage facility (Inventory). Ideally radiological characterisation is dependant on a strict system of WAC to ensure that the engineers think about the overall process. As no final WAC have been formulated DD needs to have a flexible approach to characterisation. DD therefore takes samples and stores them for the future.

Radiological characterisation of key nuclides is needed during decommissioning and dismantling. The information gained from the characterisation helps in the planning phase prior to the dismantling and can be used for inventory calculations later. Due to this DD have standard procedures for performing the radiological characterisation via both non-destructive and destructive analysis on samples.

DD has found that sampling yields more precise results than models. The variable nature of the decommissioned waste means that the activity distribution of the waste is also very varied across systems.
An example is the biological shield of DR2. The shield was made of barytes concrete and therefore contained Ba-133 among other induced isotopes. Sampling by drilling was done to investigate if the outer part of the concrete could be free released. The samples showed differences in the activity concentration of Ba-133 in the inner part of the concrete closer to the core. A sketch of the samples in the biological shield can be seen in Figure 3.

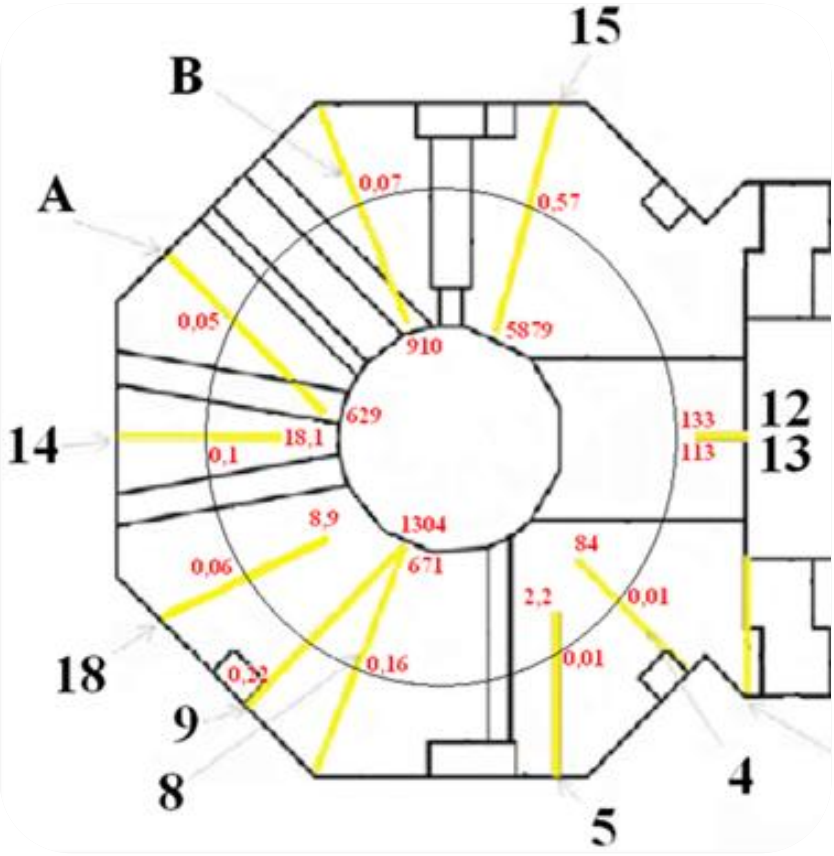

Figure 3, Sketch of drilling holes in the biological shield of DR2. The biological shield was made of barytes concrete. The red numbers are measured activity concentrations of Ba-133.

The concrete waste was put into several containers. The samples proved more effective in differentiating between the containers in the inventory calculation than a model based on symmetry would be.

Inventory

The samples are measured with gamma spectroscopy using mathematical and geometrical analysis. In the inventory calculations scaling factors are used for neutron activated waste (DR1, DR2 and DR3) to determine the difficult-to-measure isotopes and pure beta emitters. The primary scaling isotope is Co-60.

The calculations are based on the cross sections, mostly the neutron capture cross section of the individual isotopes being subject to the neutron fluence. The calculations are therefore also based on the trace elements of the irradiated material. Lastly the

calculations are dependent on an irradiation time and a decay time which are unique to each nuclear facility.
The formulas used calculate the induced activity per grammes of target after an irradiation time of *tirr* years and a decay time of *tdecay* years:

Table 1, Formula for calculating the induced activity per grammes of target after an irradiation and a decay time.

$$q(tirr, tdecay) = \frac{N}{A} \cdot \frac{a}{100} \sigma \cdot \phi \cdot \left(1 - \exp\left(\frac{-\ln(2)}{thalf} \cdot tirr\right)\right) \cdot \exp\left(\frac{-\ln(2)}{thalf} \cdot tdecay\right)$$

Where *N* is Avogadros number, *A* is grammes/mol of the target nuclide, *a* is the abundance of the target isotope in percent, σ is the reaction cross section, ϕ is the thermal fluence rate, *thalf* is the half life of the isotope, *tirr* is the irradiation time, *tdecay* is the decay time.
The term outside the paranthesis describes the amount of target isotopes capturing the incoming neutrons. The first paranthesis is a term describing the decay during irradiation and the last paranthesis is the decay after shutdown of the reactor.

It is difficult to calculate the thermal fluence rate. By ignoring the the constants the formula can be used for relative scaling factors with respect to each other.

Table 2, Formula for calculating scaling factors for the individual reactions.

$$SF(tirr) = \frac{a}{A} \cdot \sigma \cdot \left(1 - \exp\left(\frac{-\ln(2)}{thalf} \cdot tirr\right)\right) \cdot \exp\left(\frac{-\ln(2)}{thalf} \cdot tdecay\right)$$

By normalising to a key nuclide, scaling factors relative to the key nuclide can be found. These can be used to calculate the activity of an isotope if the activity of the key nuclide such as Co-60 is known. These scaling factors are unique for each nuclear facility with a specific irradiation time and decay time, each irradiated material with a specific amount of trace elements and each isotope to be scaled. This information must be known when applying the scaling factors.
Scaling factors do not take contamination into account, however if the contamination consists of neutron irradiated products such as rust, they are applicable. If this is not the case other methods must be taken into account.
For example waste from the Hot Cell facility is alfa contaminated due to transuranides and uranium experiments. In order to use scaling factors for the Hot Cell facility, DD has measured samples and on this basis calculated scaling factors where the key nuclide is Cs-137. This has the benefit of not being dependent of the contaminated material.
Examples of scaling factors for irradiated steel from DR3 and contaminated waste from Hot Cell is shown in Table 3.

Table 3, Scaling factors for common nuclides for neutron activated DR3 steel and Hot Cell contamination.

Activation - Isotopes	DR3 Steel: Co-60 Scaling factors
Ni-63	12,01
Fe-55	6,067
Eu-152	2,792
Ba-133	0,4873

Contamination - Isotopes	Hot Cell: Cs-137 Scaling factors
Pu-241	1,574
Sr-90	0,535
Pu-238	0,114
Am-241	0,093

Cl-36	0,3814
Eu-154	0,1497
Ni-59	0,1270

Pu-239	0,054
Pu-240	0,041
Cm-243/Cm-244	0,035

The scaling factors for neutron activated waste are calculated from the neutron capture cross sections for each isotope and measured trace elements in the different materials. The scaling factor for an isotope for a material is multiplied to a measured Co-60 activity to calculate the difficult-to-measure isotopes. For Hot Cell waste the scaling factors are determined from sampling and measurements. The scaling factors are not dependend on the waste material as the activity is in the contamination. The primary scaling isotope for Hot Cell waste is Cs-137 as it can easily be measured with γ-spectroscopy. The primary scaling isotopes are measured through representative sampling of the waste systems.

Ad Hoc Sampling

Due to the variable nature of the systems to be decommissioned DD uses a flexible approach to sampling for radiological characterisation for planning and for inventory calculations. A very general standardized waste flow model for DD is shown in Figure 4.

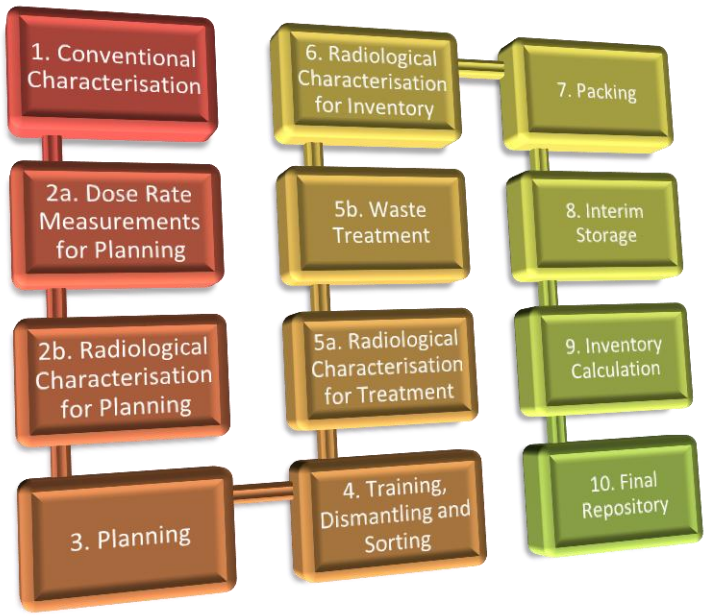

Figure 4, Standardized waste flow. An Ad Hoc approach.

In order to dismantle a system a work plan must be written. This is the Planning phase (3). Ideally the planning phase is based on conventional and radiological information about the system to be dismantled. If no such information exists it must be generated (1 and 2). Conventional characterisation is very important (1).

For example the decommissioning of the Hot Cell facility faces problems about lack of documentation of previous work done in the hot cells during operation. Several hot spots are present inside the cells as Cobalt and uranium pellets was dropped onto the floor during the usage of the facility. When the facility was closed down the walls were washed in only some of the cells and the debris were brushed into piles with brooms. These piles were never documented and several hot spots are contained in each pile. Lack of conventional and radiological characterisation tends to bring unforseen problems later.

This examples show that the more knowledge about a system exists beforehand, the less new information will show up later. Thorough conventional characterisation is very important and can sometimes eliminate the need to do radiological characterisation (2) before the planning step (3). This ensures added flexibility in the sampling as it can be done later in the process (5 or 6) and hopefully more representatively for the resulting waste.
The first step of the approach should therefore be to collect as much conventional knowledge about the system as possible. One should be aware that old documentation can be wrong, as improvements to the system could have been implemented after the documentation was made. Rarely new documentation was made when systems were improved.

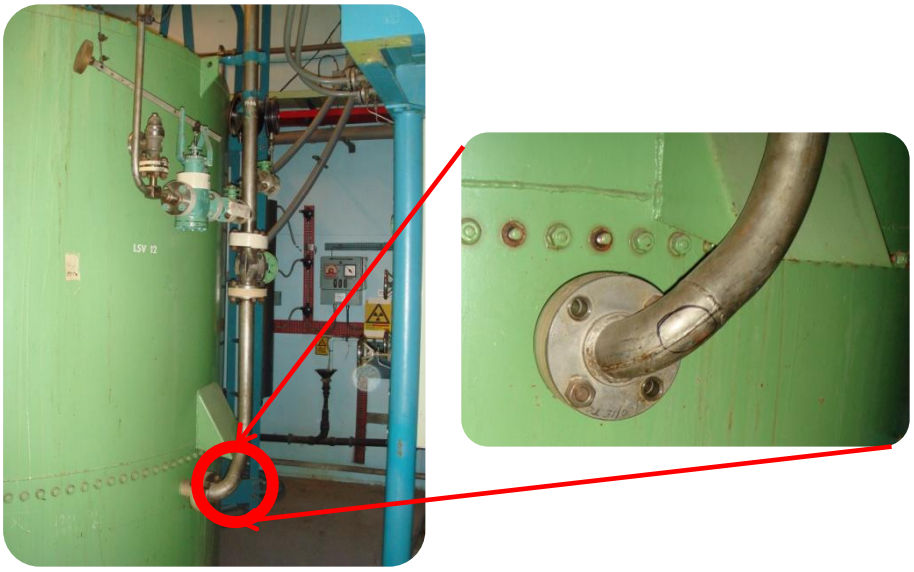

Figure 5, Example of pictures from a work plan. The place to sample is clearly marked. The sampling will be done prior to dismantling of the system.

Sampling can be done before dismantling and sorting as in Figure 5. In this case the precise origin of each waste item is well known and it is therefore easy to sample representatively.
Care must be taken to ensure the documentation and representability of the samples. In Figure 6, a dismantled and sorted system is shown. The sampling took place prior to the dismantling and the samples can be referenced to each sorting box.

Figure 6, Dismantled system sorted in materials. Sampling has taken place prior dismantling.

One must decide when the sampling should take place. Knowledge about the activity koncentration is sometimes needed in the planning phase. Radiological characterisation should then be done before. Otherwise sampling should be between the dismantling of the system and the packaging of the final containers. In this way it will be ensured that the samples are representative of the dismantled system and the packaged containers.

Samples can be stored for the future to enable a new characterisation. Representative sampling and documentation then becomes even more important.
Denmark has no WAC, so DD stores samples for the future. It is then important to be very ambitious and do the sampling as thoroughly as possible. In practice sampling takes time away from the practical dismantling work and can become a problem if high doses are involved. This increases the demands for the planning of the decommissioning task.

There is no general best standard method as described in Figure 6. The best method for one system is different from the best method for another system.
The favorable approach is therefore to determine what the best method is for the system in question. This approach is very ad hoc and involves looking at the decommissioning process as a whole. In order to cover all angles several people with different expertises need to be involved.

Lately DD has focused more on waste minimization. When external contractors will be used in the future, our procedures will need to be changed to accommodate the external contractors WAC.
DD is currently implementing a new waste policy. This policy encourages cradle-to-grave outlook and involvement of all expertises early in the process.

Summary

Nuclear facilities consist of a huge variety of parts which turns into heterogeneous waste when decommissioned. Decommissioning is project oriented where unique never seen before problems occur. This needs to be taken into account in the planning phase (in the ideal world it is done before the commissioning). A flexible Ad Hoc approach to

decommissioning is therefore invaluable. The approach should be based on cradle to the grave principles where the whole process is taken into account.
Sampling procedures should be implemented in the planning phase to ensure that the whole process is being looked at. Depending on the system to be decommissioned the representative sampling step in the process can be done as a part of the planning or during the dismantling. The sampling should not be done a long time after the dismantling, or the representativeness of the system will be lost.

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