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# Validation of activity determination codes and nuclide vectors

Workshop on Radiological Characterisation for Decommissioning
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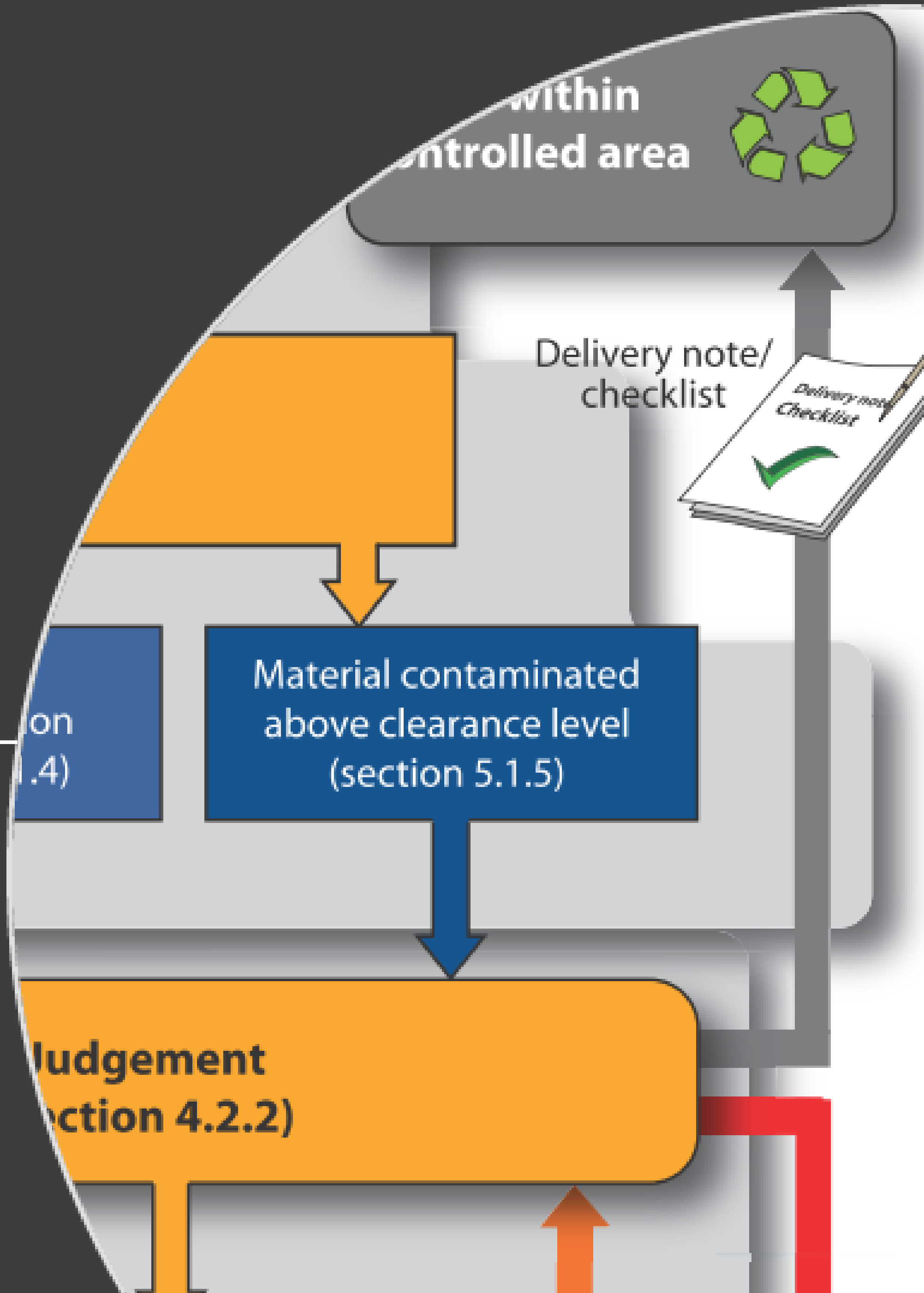
# Background

- Studsvik was founded in 1947 as a state own research company with  the mission to develop nuclear power in Sweden.
- Today Studsvik is a private commercial company focused on providing high quality services to the domestic and international nuclear industry.
- Already in the old days Studsvik paid a lot of attention in materials characterisation. The interest in characterisation of waste and in characterisation for decommissioning has increased over time.
- Studsvik has built several theoretical and practical references from characterisation of materials, systems and buildings for decommissioning.

# Introduction

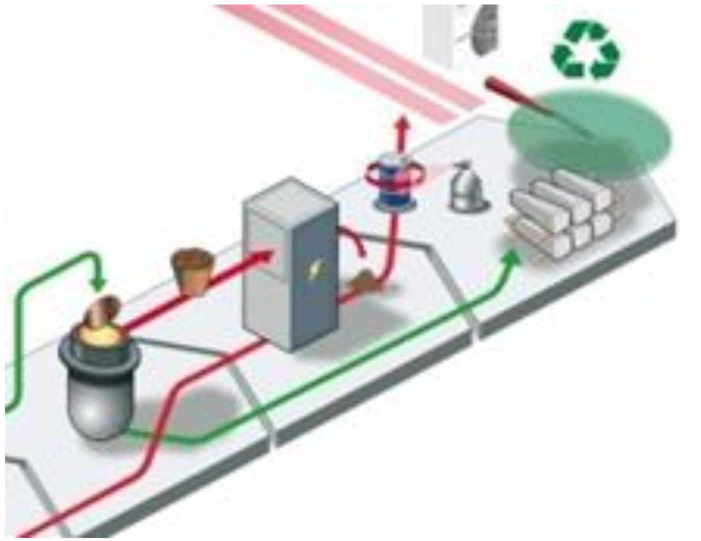


# Initial facts

- All radioactive or potentially radioactive objects need to be characterised - either for clearance or disposal
- Critical parameters for characterisation differs between routes. Has to be considered already in the validation planning
- Characterisation for clearance is easier than characterisation for disposal
- Quality assurance of activity assessment calculations and nuclide vectors requires validation require physical measurements
- 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





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# Areas with development potentials

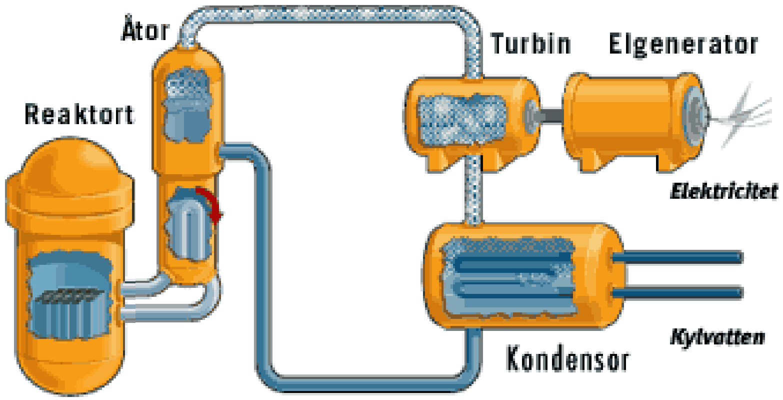
- Validation of calculation models for hard to measure nuclides.
- Validation of activity assessment calculations for systems with low contamination levels far from the reactor grid.

- 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

# Importance of good understanding

Process parameters:

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



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# 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 behavior 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

| | | | | V |
|---|---|---|---|---|
| | | E+09 | 2.4E+09 | 4.5E+0 |
| | | 1.0E+02 | 3.6E+05 | 6.6E+0 |
| | 09 | | | |
| | 3E+13 | 2.9E+07 | 4.7E+08 | 2.1E+0 |
| | 9.0E+15 | 7.6E+09 | 6.4E+10 | 4.2E+0 |
| | 2.3E+15 | 1.0E+10 | 1.8E+11 | 4.5E+0 |
| | 2.2E+13 | 4.0E+07 | 8.0E+08 | 1.9E+0 |
| | 2.8E+15 | 5.2E+09 | 9.5E+10 | 2.5E+0 |
| 90 | 5.6E+08 | 1.3E+08 | 6.1E+08 | 1.1E+0 |
| -94 | 3.5E+10 | 8.3E-03 | 3.3E+00 | 6.2E-0 |
| c-99 | 5.1E+10 | 2.2E+04 | 1.2E+07 | 2.3E+0 |
| Sb-125 | 6.7E+11 | 2.3E+08 | 2.1E+09 | 1.8E+0 |
| I-129 | 1.5E+05 | 6.1E+03 | 5.5E+04 | 1.0E+0 |
| Cs-134 | 3.9E+09 | 6.7E+04 | 1.8E+09 | 3.3E+0 |
| Cs-135 | 6.9E+05 | 3.8E+04 | 3.7E+05 | 6.8E+0 |
| Cs-137 | 9.4E+10 | 5.7E+08 | 2.6E+10 | 4.9E+0 |
| Eu-152 | 2.0E+11 | | | |
| Eu-154 | 1.4E+10 | 4.1E+04 | 2.2E+07 | 4.2E+0 |
| Eu-155 | 5.2E+09 | 1.0E+04 | 6.6E+06 | 1.2E+0 |
| Pu-238 | 1.5E+07 | 4.3E+04 | 1.7E+07 | 3.3E+0 |
| Pu-239 | 7.0E+06 | 8.0E+03 | 3.1E+06 | 6.0E+0 |
| Pu-240 | 3.9E+06 | 1.3E+04 | 5.1E+06 | 9.7E+0 |
| Pu-241 | 3.0E+08 | 6.5E+05 | 2.6E+08 | 5.4E+0 |
| Am-241 | 8.7E+06 | 2.8E+04 | 1.0E+07 | 1.9E+0 |
| m-244 | 1.0E+07 | 2.4E+04 | 9.5E+06 | 2.0E+0 |
| [kg] | 2.3E+06 | 1.2E+06 | 3.1E+05 | 2.1E+0 |

# Determination of decommissioning activity inventories in reactors – Reference list

- **Olkiluoto 1 and 2**
  - Process systems, database modification and update (2008)
- **Barsebäck 1 and 2**
  - 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**
  - Total activity assessment (2007), updates (2010, 2012)
- **Forsmark 1, 2 and 3**
  - Total activity assessment (2010), update (2012)
- **Oskarshamn 1, 2 and 3**
  - Total activity assessment (2010), update (2012)
- **Ågesta (PHWR, closed in 1974)**
  - Total activity assessment (2010)

# Source terms considered

- **Neutron induced activity in internals, RPV and biological shield (insulation, concrete and reinforcement)**
  - Neutron flux (MCNP), material compositions, neutron activation calculations (IndAct, FISPACT)
- **Activated corrosion products on system surfaces ("Crud")**
  - Model CrudAct for BWR and PWR for primary system
  - Relative distribution between different systems based on dose rate and gamma scan measurements
- **Fission products and actinides from leaking fuel:**
  - SAR leakage models combined with plant data
  - Fuel dissolution results in:
    - Actinide incorporation in system oxides (long memory effect)
    - Tramp U on core → Noble gas daughters in offgas systems
- **Contamination of concrete from system leakage**

# Nuclides considered

- **Initially 28 nuclides:**
  - 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
- **Further 20 nuclides included in 2012 update:**
  - Be10, Se79, Mo93, Zr93, Nb93m, Ru106, Pd107, Sn126, Ba133, Pm147, Ho166m, U232, U236, Np237, Pu242, Am242m, Am243, Cm243, Cm245, Cm246
- **Some additional nuclides being discussed, e.g.:**
  - Fe60 ($T_{½}$ = 1.5 My)
    - Fe58 (n, $\gamma$) Fe59 (n,$\gamma$) **Fe60** ($\beta$-) Co60m (IT) **Co60**
      Fe60 has high dose factors both for ingestion and inhalation
- **Additional nuclides call for improved evaluation and validation processes**



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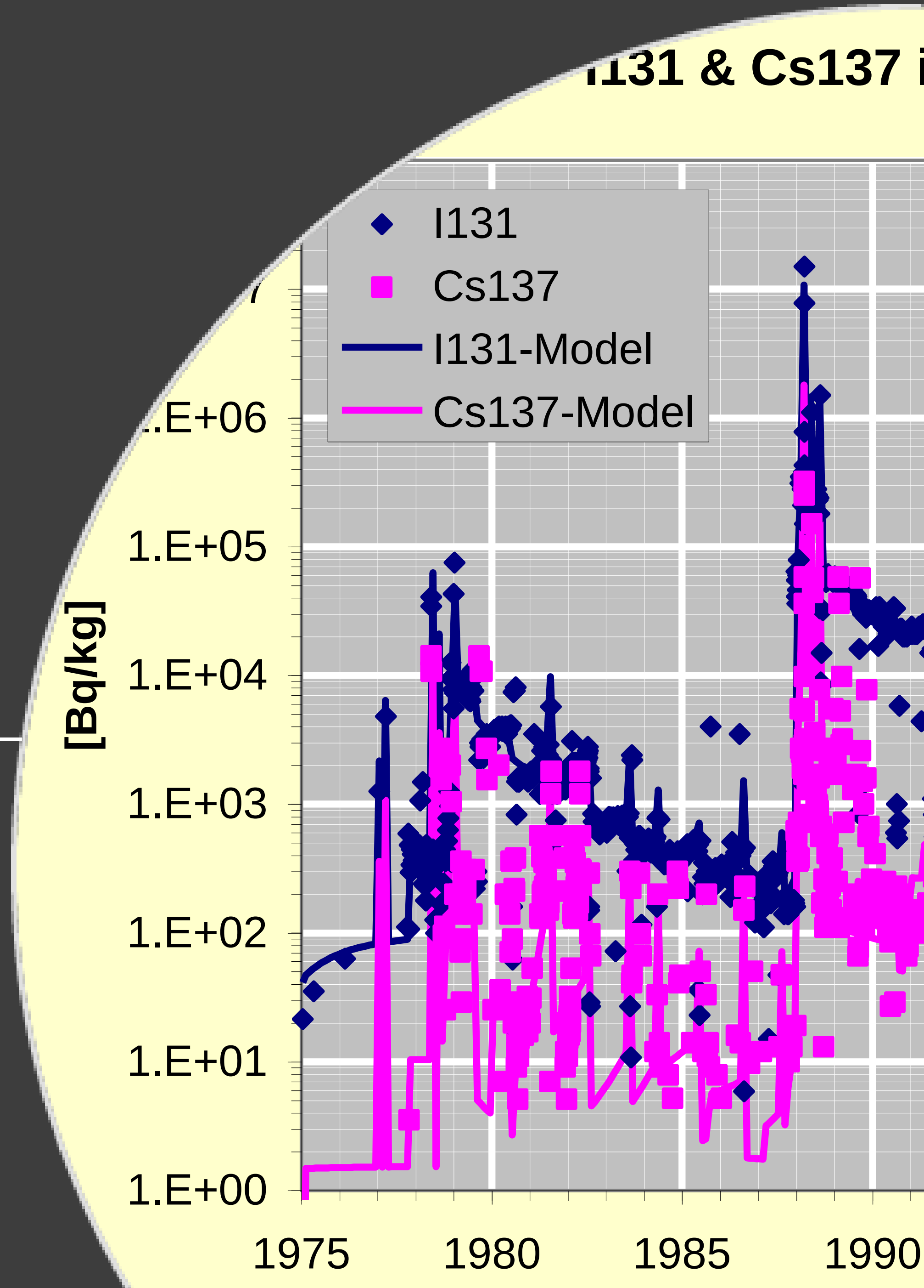
# Recent calculation results for typical BWR fuel crud – FISPACT-2007 – Ingestion and inhalation dose after decay 1000 y

- Specific activity (Bq per gram fuel crud) has been calculated
- A decay period of 1000 y has been considered, and the activity has been recalculated to ingestion and inhalation doses per gram fuel crud
  - **Fe60 is on the Top10-list**
- **NOTE: The transport properties in the environment is not considered**

| 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 performed validations



# *Example of validation:*
# Ringhals-3 Steam Generator

- **Higher waste activity than determined by in-plant gamma scanning**
  - Complicated geometry to evaluate gamma scans (only peripheral tubes)
- **Ni63/Co60 ratio in line with earlier assessments**
- **C14 was detected in the blasting waste**
  - The detected amount corresponds to about 0.24% produced in the coolant during one year
  - Additional evaluation initiated

**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 |

**Ni63 measurements performed by Studsvik**
**C14 measurements performed by Ringhals AB**

*Example of validation:*

# B1 & 2 – Activity removed from system decontaminations

| 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)**

*Example of validation:*

# 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 validation:* F1/2/3 –Turbine components

- **Totally about 1300 tons of turbine components**
  - About 95% subject to clearance, 71% direct clearance
  - Remaining 5% is waste for disposal
- **Totally about 1.6 GBq Co60**
  - In line with assessment calculations
- **Totally about 0.09 GBq Cs137**
  - Higher than assessment calculations
  - TU → Xe137 (3.8 min) → Cs137
    - Dominating fraction expected in off-gas systems
- **Other nuclides detected**
  - Mn54, Co60, Zn65, Ag110m

| | | 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 and the way forward

# Conclusions

- 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
- Joint efforts can make change

# A way forward

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.

- Open books inside project – joint financing - common learning
- Use retired components sent for treatment as objects for validations
- Special areas of interest
  - long lived beta-emitting nuclides
  - nuclide distribution, behavior and effects of treatment
  - optimisation of sampling and evaluation

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