DIFFICULT TO MEASURE BETA EMITTERS (\(^{55}\text{Fe}\) AND \(^{63}\text{Ni}\)) IN ACTIVATED PRESSURE VESSEL STEEL – THEORETICAL VERSUS EXPERIMENTAL ANALYSIS

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• Radionuclide distribution in decommissioning waste has to be determined prior to dismantling with radiochemical separations for determining beta and alpha emitters (DTM radionuclides)
  -> Material-dependent nuclide vectors from this experimental data
  -> After dismantling the decom materials can be characterized with simple gamma spectrometric measurements combined with nuclide vectors

• Real activated RPV steel as a sample matrix gives more realistic view than simulated experiments with inactive steel and radioactive tracers

• Beta emitters have to be purified from other beta and gamma emitters (e.g. $^{60}$Co) before activity determination by LSC

• Method for large amount of dismantling waste: Precise but slow determination method for few samples vs. coarser but faster method for more samples?
RADIOCHEMICAL SEPARATION OF $^{55}\text{Fe}$ AND $^{63}\text{Ni}$ FROM STEEL

• Different separation methods were tested with inactive and activated steel
• The separation methods included hydroxide co-precipitation of metals, different anion exchange schemes, purification of Ni with Ni resin ® or DMG co-precipitation
• High concentration of $^{60}\text{Co}$ in steel samples was problematic with all separation methods
• Open questions after our pilot project:
  * acceptable level of $^{60}\text{Co}$ in the separated fractions of $^{55}\text{Fe}$ and $^{63}\text{Ni}$
  * high recovery of $^{55}\text{Fe}$ and $^{63}\text{Ni}$ with slow method vs. lower recoveries with faster method
LSC spectra of $^{55}$Fe separated from steel before (left) and after (right) extra anion exchange step performed in acetone + HCl mixture.
RADIOMETRIC MEASUREMENTS

$^{60}$Co by ISOCS (In-Situ Object Counting System)

- the counting efficiency of a particular measurement comprises of the sample size, thickness and density of the sample and its container, collimators if present, and the source-detector distance

$^{60}$Co by standard geometry calibration: GX 8021 HPGe spectrometer (Canberra)

- steel sample in an LSC vial of 20 ml
- Genie 2000 Gamma Acquisition & Analysis program (Canberra)
- Dual polynomial fitting option was used for efficiency and energy calibration

- $^{55}$Fe and $^{63}$Ni by LSC (Quantulus 1220 and Hidex 300 SL)
COMPUTATIONAL METHODS

- The calculations were performed using a point kinetic code **ORIGEN-S** (I.C. Gauld et al., ORIGEN-S: A Scale System Module to Calculate Fuel Depletion, Actinide Transmutation, Fission Product Buildup and Decay, and Associated Radiation Source Terms, ORNL/TM-2005/39, version 6.1, 2011)

- theoretical activity concentrations of the radionuclides in the activated steel based on manufacturing specifications and known irradiation history
## RESULTS: $^{60}$Co IN BWR STEELS (UNCERTAINTY OF A ~1-3%)

<table>
<thead>
<tr>
<th>Sample</th>
<th>$^{60}$Co by ISOCS (Bq/g)</th>
<th>Experimental/theoretical by ISOCS</th>
<th>$^{60}$Co by standard calibration (Bq/g)</th>
<th>Experimental/theoretical by stand. cal.</th>
</tr>
</thead>
<tbody>
<tr>
<td>BWR_a</td>
<td>16 300</td>
<td>0.9</td>
<td>16 300</td>
<td>0.9</td>
</tr>
<tr>
<td>BWR_b</td>
<td>15 900</td>
<td>0.8</td>
<td>14 600</td>
<td>0.8</td>
</tr>
<tr>
<td>BWR_c</td>
<td>15 800</td>
<td>0.8</td>
<td>13 700</td>
<td>0.7</td>
</tr>
<tr>
<td>BWR_d</td>
<td>16 000</td>
<td>0.9</td>
<td>15 900</td>
<td>0.9</td>
</tr>
<tr>
<td>BWR_e</td>
<td>16 200</td>
<td>0.9</td>
<td>n/a</td>
<td></td>
</tr>
<tr>
<td>BWR_f</td>
<td>15 800</td>
<td>0.8</td>
<td>n/a</td>
<td></td>
</tr>
<tr>
<td>BWR_g</td>
<td>16 000</td>
<td>0.9</td>
<td>n/a</td>
<td></td>
</tr>
<tr>
<td>BWR_h</td>
<td>16 200</td>
<td>0.9</td>
<td>n/a</td>
<td></td>
</tr>
</tbody>
</table>
## RESULTS: $^{60}$Co in VVER Steels

(UNCERTAINTY OF A ~1-3%)

<table>
<thead>
<tr>
<th>Sample</th>
<th>A $^{60}$Co by ISOCS (Bq/g)</th>
<th>Experimental/theoretical by ISOCS</th>
<th>A $^{60}$Co by standard calibration (Bq/g)</th>
<th>Experimental/theoretical by stand. cal.</th>
</tr>
</thead>
<tbody>
<tr>
<td>VVER_a</td>
<td>68 500</td>
<td>0.9</td>
<td>71 100</td>
<td>1.0</td>
</tr>
<tr>
<td>VVER_b</td>
<td>71 400</td>
<td>1.0</td>
<td>69 900</td>
<td>0.9</td>
</tr>
<tr>
<td>VVER_c</td>
<td>70 100</td>
<td>0.9</td>
<td>71 600</td>
<td>1.0</td>
</tr>
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<td>VVER_d</td>
<td>72 200</td>
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RESULTS: $^{55}$Fe AND $^{63}$Ni IN BWR STEELS (UNCERTAINTY OF A ~14%)
RESULTS: $^{55}$Fe AND $^{63}$Ni IN VVER STEELS (UNCERTAINTY OF A $\sim$14%)
CONCLUSIONS

• Differences in chemical composition between two steel types -> differences in chemical behaviour during separation procedure, and radionuclide distribution

• both gamma calibration approaches (ISOCS and standard calibration) worked equally well for $^{60}$Co, but for weaker gamma energies this would probably not have been the case

• Experimental/theoretical values for
  
  $A^{^{60}}$Co: 0.7-1.0  
  $A^{^{55}}$Fe: 0.7-2.0  
  $A^{^{63}}$Ni: 0.6-1.9

  -> overall: experimental and theoretical values correlate pretty well

• The current situation of the pilot project: evaluation of uncertainties and writing a manuscript
FORTHCOMING ARTICLE

• A Master thesis was produced during this project by Taneli Iso-Markku, UH: "Difficult-to-measure beta active radionuclides in nuclear decommissioning waste"

• Anumaija Leskinen, Susanna Salminen-Paatero, Merja Tanhua-Tyrkkö, Taneli Iso-Markku, Esa Puukko, Antti Räty:

“Determination of $^{14}$C, $^{55}$Fe, $^{63}$Ni and gamma emitters in activated RPV steel samples - a comparison between calculations and experimental analysis”

• TO BE SUBMITTED TO JOURNAL OF RADIOANALYTICAL AND NUCLEAR CHEMISTRY IN SUMMER 2019

-> Stay tuned!
CONTINUANCE: THE "DEMONI" PROJECT, 2019-2023

• Funded by KYT2018 Research program
• To expand the selection of nuclear decommissioning materials analyzed and validate the separation methods
• Matrices: steel, concrete, spent ion exchange resin
• Radionuclides: $^{14}\text{C}$, $^{60}\text{Co}$, $^{55}\text{Fe}$, $^{63}\text{Ni}$, $^{41}\text{Ca}$, others (?)
• Also long-term dissolution tests of steel in ground water (UH and VTT)
• Mechanical properties of concrete and dissolution behavior of concrete in conditions simulating final disposal of nuclear waste (VTT)
• Will be executed in co-operation and co-funding with NKS projects, e.g. ongoing DTM-DECOM project (intercomparison in determination of fore mentioned radionuclides in activated pressure vessel steel)
ACKNOWLEDGEMENTS

- KYT 2018 (Finnish Research Programme on Nuclear Waste Management 2015-2018)
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THANK YOU FOR LISTENING!