DIFFICULT TO MEASURE BETA EMITTERS (^{55}FE AND ^{63}NI) IN ACTIVATED PRESSURE VESSEL STEEL – THEORETICAL VERSUS EXPERIMENTAL ANALYSIS

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BACKGROUND AND MOTIVATION

- 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?

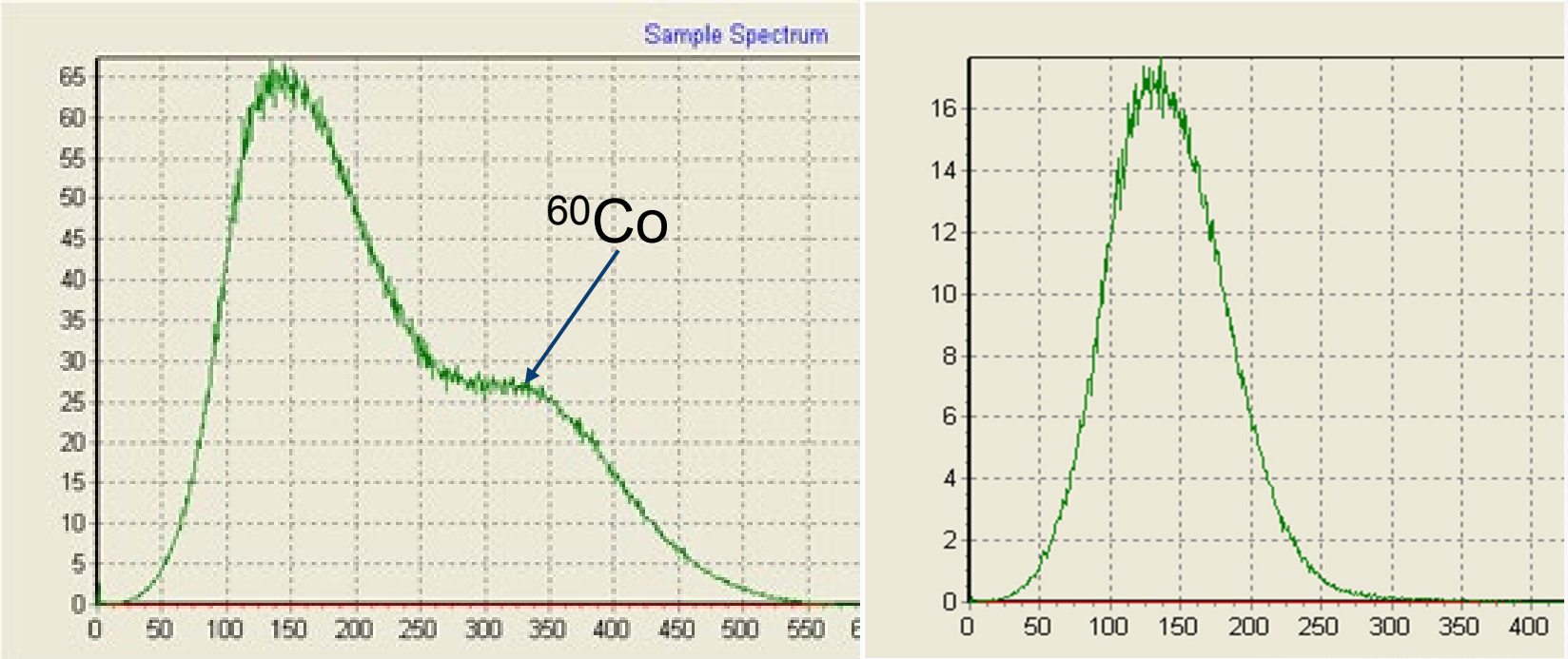
RADIOCHEMICAL SEPARATION OF ^{55}FE AND ^{63}NI FROM STEEL

- 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}Co in steel samples was problematic with all separation methods
- Open questions after our pilot project:

  * acceptable level of ^{60}Co in the separated fractions of ^{55}Fe and ^{63}Ni

  * high recovery of ^{55}Fe and ^{63}Ni with slow method vs. lower recoveries with faster method

^{60}CO CONTAMINATION IN SEPARATED FE FRACTION



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%)

Sample	A ^{60}Co by ISOCS (Bq/g)	Experimental/ theoretical by ISOCS	A ^{60}Co by standard calibration (Bq/g)	Experimental/ theoretical by stand. cal.
BWR_a	**16 300**	**0.9**	**16 300**	**0.9**
BWR_b	**15 900**	**0.8**	**14 600**	**0.8**
BWR_c	**15 800**	**0.8**	**13 700**	**0.7**
BWR_d	**16 000**	**0.9**	**15 900**	**0.9**
BWR_e	16 200	0.9	n/a	
BWR_f	15 800	0.8	n/a	
BWR_g	16 000	0.9	n/a	
BWR_h	16 200	0.9	n/a	

RESULTS: ^{60}CO IN VVER STEELS (UNCERTAINTY OF A ~1-3%)

Sample	A ^{60}Co by ISOCS (Bq/g)	Experimental/ theoretical by ISOCS	A ^{60}Co by standard calibration (Bq/g)	Experimental/ theoretical by stand. cal.
VVER_a	**68 500**	**0.9**	**71 100**	**1.0**
VVER_b	**71 400**	**1.0**	**69 900**	**0.9**
VVER_c	**70 100**	**0.9**	**71 600**	**1.0**
VVER_d	**72 200**	**1.0**	**68 300**	**0.9**
VVER_e	72 800	1.0	n/a	
VVER_f	71 900	1,0	n/a	
VVER_g	72 000	1.0	n/a	
VVER_h	72 000	1.0	n/a	

RESULTS: ^{55}FE AND ^{63}NI IN BWR STEELS (UNCERTAINTY OF A ~14%)

Sample	A ^{55}Fe (Bq/g)	A ^{55}Fe meas/calc	A ^{63}Ni (Bq/g)	A ^{63}Ni meas/calc
BWR_a	278 200	2.0	6 000	1.1
BWR_b	224 300	1.6	6 400	1.1
BWR_c	128 200	0.9	6 100	1.1
BWR_d	123 400	0.9	5 900	1.1
BWR_e	228 400	1.6	3 600	0.6
BWR_f	99 200	0.7	N/A	N/A
BWR_g	243 200	1.7	5 700	1.0
BWR_h	217 700	1.5	5 900	1.1

RESULTS: ^{55}FE AND ^{63}NI IN VVER STEELS (UNCERTAINTY OF A ~14%)

Sample	A ^{55}Fe (Bq/g)	A ^{55}Fe meas/calc	A ^{63}Ni (Bq/g)	A ^{63}Ni meas/calc
VVER_a	135 400	1.4	52 600	1.0
VVER_b	118 200	1.2	59 100	0.9
VVER_c	125 800	1.3	76 100	1.3
VVER_d	122 400	1.2	74 300	1.3
VVER_e	144 400	1.5	109 200	1.9
VVER_f	141 800	1.4	101 800	1.8
VVER_g	137 600	1.4	69 900	1.2
VVER_h	147 300	1.5	64 700	1.1

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}C, ^{60}Co, ^{55}Fe, ^{63}Ni, ^{41}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)

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THANK YOU FOR LISTENING!