

Considering decommissioning waste in new reactor materials selection

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Background

- Reactor structural materials selection historically:
 - Mechanical requirements (strength, fracture toughness, fatigue)
 - Fabricability (formability, weldability)
 - Environmental stability (resistance to heat, corrosion, radiation)
 - Suitability for non-destructive examination
 - COST.
- Operational experience has resulted in some replacement choices.
- Lack of spare parts supply chain is leading to exploration of additive manufacturing as an alternative means of fabrication.

Background

- In nuclear reactor building boom of 70s and 80s, focus was on power production; driven by cost.
- But materials can **activate** under neutron irradiation!
- Impact of activation products on inspections and dismantling *has* given rise to e.g. replacement of cobalt-based alloys (Stellites).
- End-of-life decommissioning and ultimate disposal **not** typically a priority factor in design and materials selection process.
- Very few, if any, materials or mechanical engineers are familiar with nuclide evolution upon exposure to neutrons.

Background

- New boom in SMR and fusion start-ups backed by hedge fund investors creates pressure to produce “results” quickly.
 - Designs are made quickly, and materials selection decisions may not be thought through very completely.
 - In some cases, adequate materials may not even exist yet, and so alloy development is still underway.
- Central to the business case for most SMRs is *mass production*.
- A “safety/security” feature of some SMRs is “*replace rather than refuel*.”
- Mass production and relatively short lifetimes means significant waste!
- Unlike an automobile, an SMR with activated materials cannot just go to the crusher!
- Engineers responsible for design and materials selection should also consider the ultimate radioactive waste resulting from their decisions!

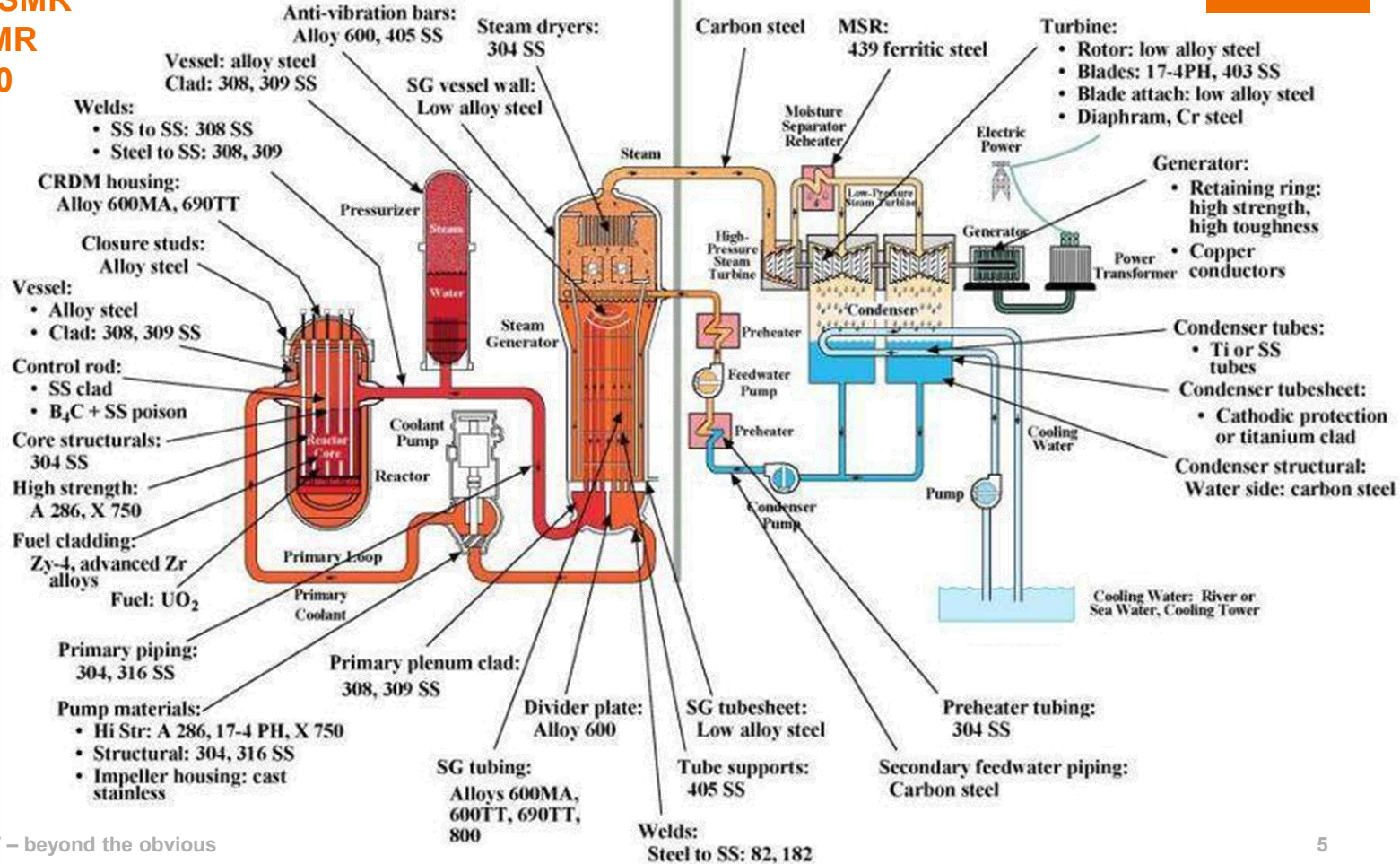
- NuScale Power Module
- Westinghouse SMR
- Rolls-Royce SMR
- Holtec SMR-160
- SMART

Primary Circuit

PWR

Secondary Circuit

VTT



Decommissioning as a design consideration

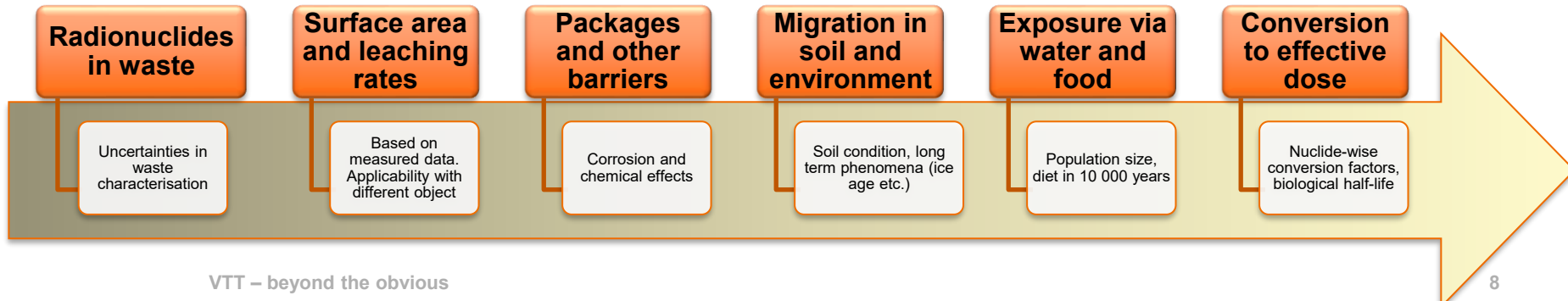
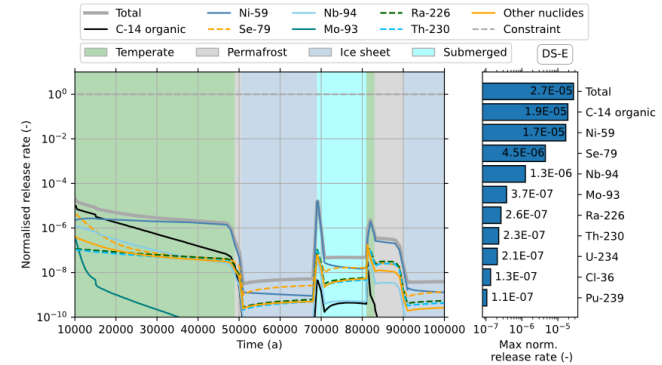
- The particular nuclides leading to activation of structural materials depend on the initial composition of the material.
- Alloy design involves major elements and minor elements.
- “Trace” or “tramp” elements originating from ores are merely minimized to below some practical economic limit.
- Besides obvious choices like avoiding Co-based materials (most high-entropy alloys!), attention needs to be paid to minor and trace elements.
- Concrete involves natural minerals in aggregate as well as cement, the compositions of which are very difficult to control in practice.

Concepts: key-nuclide, DTM nuclide, nuclide vectors

- As materials activate during reactor operation, radionuclides produced can be divided into gamma active "key" nuclides, and beta-active difficult-to-measure (DTM) nuclides.
- Major γ -emitters can be easily measured through γ -spectrometry (NDA technique). E.g. Co-60, Ba-133, Cs-137, Eu-152 and Eu-154.
- Minor γ -emitters may require separation from major ones before measuring is possible.
- Minor γ -emitters and DTM nuclides need dedicated sample preparation and laborious destructive measurement techniques. E.g. C-14, Cl-36, Ni-63, etc.
- A nuclide vector describes the ratios of nuclide activities in a material.
 - Fixing the nuclide vector enables smooth waste package characterization when classifying waste (both operational and dismantling)
- A nuclide vector is not constant in time; nuclear decay changes the ratios.
 - In old samples, most gammas have decayed, but DTM nuclide activities may be almost unchanged.
 - High or low gamma dose rate does not directly correspond to long-term safety in final disposal, but DTM activity does.

Long-term safety of final disposal

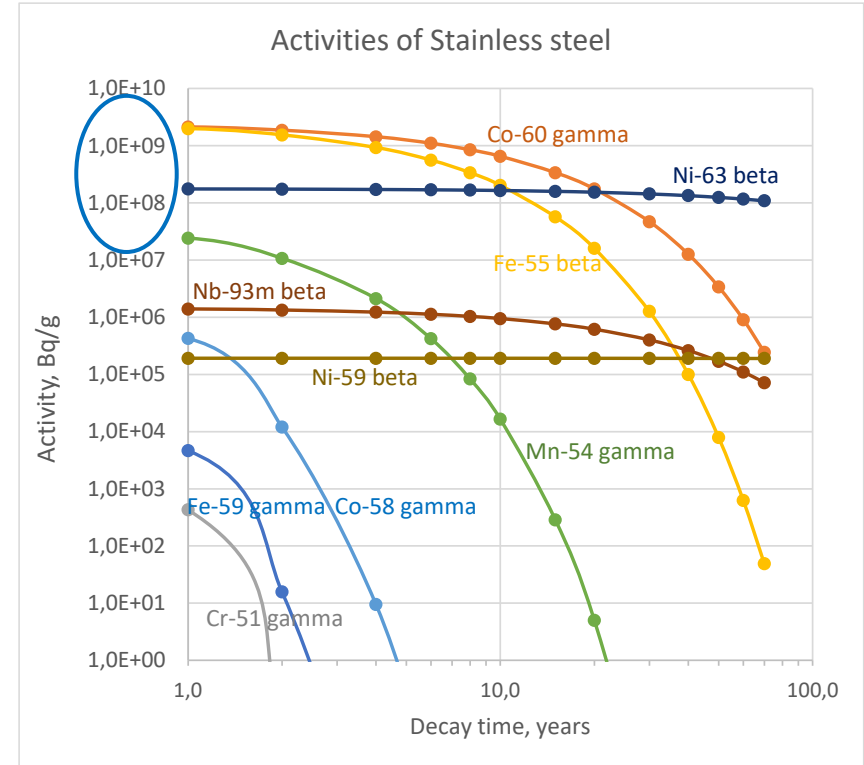
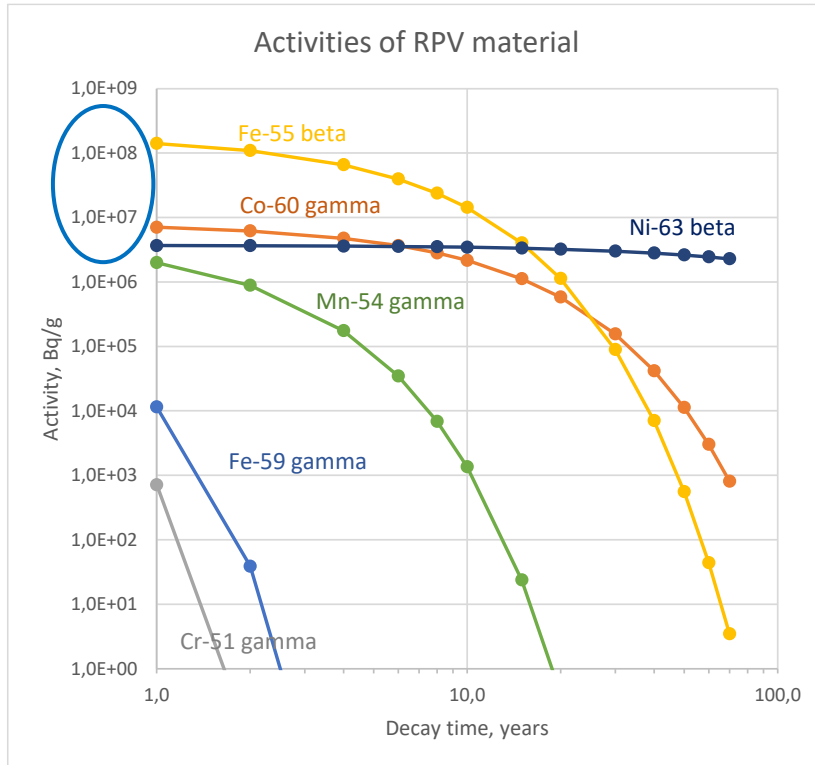
- Nuclide-wise activities only provide a source term that is used to estimate the long-term safety of waste final disposal.
- Other things to consider are e.g. leaching rate, migration, exposure and radiotoxicity
 - In addition, the chemical behaviour and long-term effects in final disposal conditions need to be taken into account.
 - Safe final disposal is dependent on other nuclides than radiation safety during dismantling!



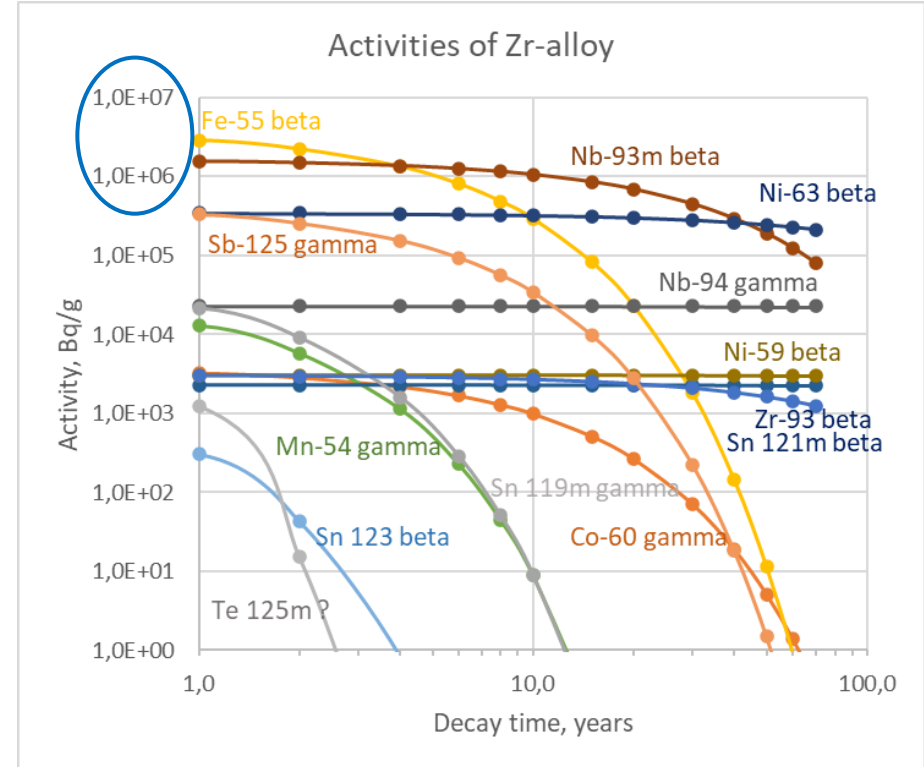
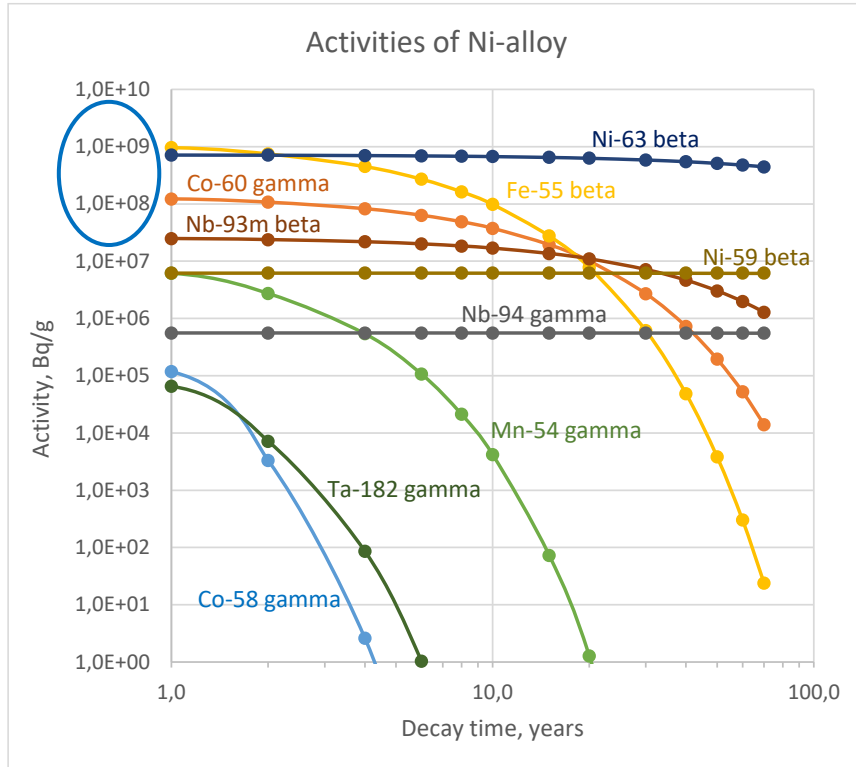
Main nuclides in activated structural materials

Isotope	Emmission	Half-life, years	A2 (Bq)	Relative proportions of nuclides in each material type, % (≠ absolute amount!)				
				RPV	Austenitic SS	Ni-base alloy	Zr-alloy	CuCrZr
Co-58	gamma	0,20	1,0E+12	0-5		<0,1		
Co-60	gamma	5,30	4,0E+11	1-12	38-47	7	<0,1	100
Fe-55	beta	2,68	4,0E+13	84	49-56	40	58	
Fe-59	gamma	0,12	9,0E+11	0-8	<0,1	<0,1		
Mn-54	gamma	0,85	1,0E+12	0-5	<0,1	<0,1	<0,1	
Nb-93m	beta	16,13	3,0E+13		<0,1	1,6	26	
Nb-94	gamma	20300	7,0E+11			<0,1	<0,1	
Ni-59	beta	108000	1,0E+99			0,4	<0,1	
Ni-63	beta	100	3,0E+13		<0,1	50	6	
Sb-125	both	2,8	1,0E+12				7	
Sn-119m	gamma	0,8	3,0E+13				1	
Ta-182	gamma	0,31	5,0E+11			<0,1		
Te-125m	gamma	0,16	9,0E+11				2	
Zr-93	both	1600000	1,0E+99				<0,1	

Decay calculations of primary nuclides



Decay calculations of primary nuclides



Calculation of nuclide vectors

Theoretical calculations for minor nuclides

- Theoretical calculations for gamma *and beta* nuclides can be made based on material composition and reactor irradiation parameters.
- Decay calculations can be made for different “cooling” periods.
- Gamma spectrometer results can be used to “calibrate” the calculations for particular materials specimens.
 - As time marches on, nuclide decay constantly changes relative amounts of nuclides in a particular material
 - For minor nuclides, accuracy may not be essential, but ***long-lived nuclides, even in small quantities, can become a disposal issue.***

Alloying elements $\neq$ most important nuclides!

Metallurgist's perspective

Steel type / element (w%)	C	Si	Mn	P	S	Ni	Cr	Mo	V	Cu	Co	Nb	N	Fe	Other
PWR RPV SA 508 Cl 2	0 - 0.25	0 - 0.4	0.5 - 1.5	0 - 0.025	0 - 0.025	0.4 - 1.0	0 - 0.45	0.45 - 0.70	0 - 0.05	0 - 0.2	0 - 0.02	0 - 0.01		Bal.	+ limits on Ca, B, Ti and Al
Austenitic SS 316L	0 - 0.03	0 - 1.00	0 - 2.00	0 - 0.045	0 - 0.015	10.00 - 13.00	16.50 - 18.50	2.00 - 2.50			0 - 0.02		0 - 0.10	Bal.	
Ni-based Alloy 718	0 - 0.08	0 - 0.35	0 - 0.35	0 - 0.015	0 - 0.015	50.00 - 55.00	17.00 - 21.00	2.80 - 3.30		0 - 0.3	0 - 1.00	4.75 - 5.50		Bal.	+ limits on Ti, Al, B
CuCrZr								0.8		Bal.					Zr: 0.08

Radiochemist's additions!

Steel type / element (w%)	Ag	Sb	Sn	Ho	Cl	Se	Zr	Zn	Pb	Al	Li	Ca	K	Ba	Eu	Cs	Sm
PWR RPV SA 508 Cl 2	0,002	0,004	0,009	0,001	0,0001	0,0009	0,0001	-	-	-	0,00003	0,0019	0,0003	0,10	0,000002	0,00007	0,00002
Austenitic SS 316L	0,0002	0,004	0,009	0,001	0,0001	0,0035	0,0001	-	-	-	0,00003	0,0019	0,0003	0,10	0,000002	0,00007	0,00002
Ni-based Alloy 718	-	-	-	-	-	-	0,0001	-	-	-	-	-	-	-	-	-	-
CuCrZr	-	-	0,002	-	-	-	0,074	0,038	0,001	0,003	-	-	-	-	-	-	-

Calculation of nuclide vectors

Theoretical calculations for minor nuclides

- Decommissioning of VTT's FiR1 reactor and structural materials hot cell involved calculating “representative nuclide vectors” for the resulting waste.
- Sensitivity study was carried out by varying reactor exposure and subsequent decay scenarios, particularly for *trace* nuclides.
 - Amount of *long-lived* nuclides in the vector is mainly a function of the initial irradiation conditions, with little change in 30 years of decay.
 - Amount of long-lived *trace* nuclides is still overall low in magnitude, but calculation error is *eclipsed by uncertainty in quantity of parent element*.
- Similar exercise described in NUREG/CR-3475, carried out by PNNL for NRC in 1984. Material activation depends greatly on
 - Reactor type (i.e. neutronics, temperature)
 - Location in the reactor core.
- SMR designs and material choices are numerous!

Calculation of nuclide vectors

Theoretical calculations for minor nuclides in RPV steel

Isotope	NUREG/ CR-3474 PWR E.O.L., Bq/g	PWR 10y irr + 15y cool, Bq/g	NUREG/ CR-3474 BWR E.O.L., Bq/g	BWR 20y irr + 20y cool, Bq/g	VVER 5y irr + 35y cool, Bq/g
C-14	2,70E+05	4,45E+03	1,81E+02	8,89E+03	2,22E+03
Cl-36	5,92E+03	1,15E+00	3,70E+00	2,29E+00	5,73E-01
Co-60	1,18E+09	7,43E+05	8,88E+05	1,71E+05	2,99E+04
Fe-55	2,18E+09	2,49E+06	1,52E+06	7,53E+05	1,20E+04
Mn-54	3,07E+07	3,94E+00	7,40E+04	6,81E-02	0,00E+00
Mo-93	2,85E+03	5,11E+01	2,85E+00	1,49E+02	3,12E+01
Nb-93m		2,41E+01		8,79E+01	2,05E+01
Nb-94	2,33E+03	1,31E+01	1,81E+00	2,62E+01	6,55E+00
Ni-59	1,59E+06	3,95E+03	1,18E+03	3,73E+03	2,08E+02
Ni-63	2,11E+08	4,22E+05	1,48E+05	3,72E+05	1,97E+04
Ag-108m	7,40E+02	7,31E+01	5,55E-01	1,38E+02	3,32E+01

Calculation of nuclide vectors

Theretical calculations for minor nuclides in NPP internals

Isotope	NUREG/ CR-3474 PWR SS Shroud E.O.L., Bq/g	PWR Type 304 Stainless steel, Bq/g	PWR Type 718 Nickel alloy, Bq/g	Generic SS 5y irr + 15y cool, Bq/g	NUREG/ CR-3474 BWR SS cladding E.O.L., Bq/g
C-14	9,25E+05	4,67E+05	2,68E+04	2,23E+03	1,81E+02
Cl-36	1,89E+04	9,53E+03	5,46E+02	5,74E-01	3,70E+00
Co-60	4,81E+09	2,43E+09	1,39E+08	1,22E+06	8,88E+05
Fe-55	7,77E+09	2,57E+09	1,24E+09	1,35E+06	1,52E+06
Mn-54	2,41E+08	5,43E+07	1,38E+07	2,69E+00	7,40E+04
Mo-93	3,48E+04	1,76E+04	1,01E+03	1,62E+01	2,85E+00
Nb-93m		1,46E+06	2,59E+07	7,30E+00	
Nb-94	1,48E+04		5,56E+05	1,46E+01	1,81E+00
Ni-59	4,07E+06	1,91E+05	6,21E+06	1,60E+04	1,18E+03
Ni-63	6,66E+08	1,77E+08	7,22E+08	1,74E+06	1,48E+05
Ag-108m	3,70E+03	1,87E+03	1,07E+02	3,71E+00	5,55E-01

Examples of "important" nuclides in waste

- This table considers only activation, not other sources of contamination. This is a simplified summary for metallic waste. List of nuclides is not universal for all waste types.
- **Minor contribution: Co-60, Fe-55, Ca-41**

Radionuclide	Activating element	Contribution
C-14	N, C	Significant
Ca-41	Ca	Minor
Cl-36	Cl	Significant
Ni-59	Ni, Co	Significant
Ag-108m	Ag	Average
Ho-166m	Ho	Minor

Radionuclide	Activating element	Contribution
Mo-93	Mo	Average
Nb-93m	Nb, Mo	Average
Ni-63	Ni	Average
Se-79	Fis. Prod.	Average
Tc-99	Mo, Fis. Prod.	Average

Summary

- Many companies are offering SMR and fusion reactor designs focused primarily on commercial energy production, driven by economics.
- Mass production is a commercial fundamental particularly for SMRs.
- Decommissioning experience has revealed that the long-term safety in the disposal of activated reactor materials may hinge more on minor and trace elements than on the obvious activation nuclides like Co-60 and Fe-55.
- Theoretical activation calculations can be used to identify problematic elements, but require knowledge of particular reactor operating conditions (neutronics) and dimensions, of which there are various in SMR sector!
- Many problematic elements are not currently typically controlled in commercial materials fabrication process, nor even economically possible to control!

Recommendations

- Safety in long-term final disposal can be increased by limiting the amount of activating elements in the materials during the construction phase.
- More attention should be paid already in the materials selection and manufacturing process phases to minimize inclusion of problematic elements.
- If this is not possible, maintain the knowledge of detailed compositions including the nuclides that may not affect to mechanical properties.
 - This data is significantly easier (and cheaper) to collect prior to starting the operation. Saving inactive reference samples of all potentially activating materials is beneficial.
 - Lesson learned from the FiR1 decommissioning project: Old facilities typically have information on mechanical properties and old documentation is seriously inadequate or can even contains false data.

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Thank you!

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