

Radionuclide inventory of burnable radioactive waste contaminated with artificial alpha-emitters

M. Rababah, K. Bauer, G. Dumont, M. Magistris, N. Menaa, P. Pisano. C. Theis

21/9/2022

Contents

- Radioactive waste at CERN
- Radioactive burnable waste
- Radiological characterization
- **BEEP-ABG Characterization**
- Conclusion



Radioactive waste at CERN

CERN's RW	Elimination pathways	
Clearance candidates - CL (Candidats à la Liberation inconditionnelle)	Release from regulatory control in Switzerland (Clearance <> "free-release")	AMAL, EVA, B-FREE, PLATAN, CRANES, etc.
Very Low-Level Waste - TFA (Très Faibles Activités)	Surface disposal in France. As defined by the acceptance criteria of the ANDRA CIRES repository	BEEP-ABG, BEEP-BG, ELICA, ELMA, SHERPA, ELFI, etc.
Intermediate & Low-Level Waste - FMA-VC (Faibles et Moyennes Activités à Vies Courtes)	Surface disposal in France Half life T _{1/2} < 30y As defined by acceptance criteria in ANDRA CSA repository	ABEILLE, MAST, etc.
Intermediate & Low-Level Waste - FA-MA (Faibles Activités et Moyennes Activités)	Disposal in Switzerland When FMA-VC acceptance criteria (half-life, activity level) are not met	STEP-1, ITEP, STEP-2, etc.

93% in volume of the waste stored at CERN belongs to the category of TFA



Burnable radioactive waste

Type of waste

- Mostly personal protection equipment
 - Gloves, fabrics, Over-shows, etc.
- ventilation filters.

BEEP-Free

• Elimination with free release in Switzerland

BEEP-BG

 Contamination by activated metallic particulates or concrete dust

BEEP-ABG

 Contamination in the manufacturing and handling of spallation targets (n_TOF) and ISOLDE







Characterization of TFA



Courtesy of B. Zaffora, M. Magistris



Challenges for predicting BEEP-ABG RN inventory

• Wide range of activation scenarios

- Targets of ISOLDE and n_TOF facilities target core and structural materials
- A sampling strategy is not adequate given the heterogeneity of the waste
 - Hence relying only on FLUKA simulation and analytical calculation

A longer list of DTMs

- Targets made of actinides produces virtually any radionuclide from nuclides table.
- Alpha emitters DTMs: Gd-148, Po-210, etc.



ISOLDE and n_TOF

• n_TOF targets

- Made of lead block
- Kept together by stainless steel frame (target1), or in aluminum (target2)
- Alpha emitters come from direct contamination
- n_TOF target #1 : 2000-2004
- n_TOF target #2 : 2008-2018

• ISOLDE targets

- Different targets materials
- Potential alpha emitters come from Uranium, Lead and Tantalum targets
- Targets are irradiated from few days up to couple of weeks





Radionuclide inventory

•	Predicting	Fluence	spectra	for the	targets -	FLUKA	simulation
---	------------	---------	---------	---------	-----------	--------------	------------

- Establishing the full list of radionuclide inventory using Actiwiz
 - For most relevant targets
 - For different irradiation times
 - For cooling times ranges from, 6 month 30 years
- Reduced radionuclide list (under certain criteria and assumptions)
 - Ex: Normalization to hazard factor IRAS = 10.
 - Half-life > 6 months
 - Excluded radionuclides (natural radionuclides): Th-232, Ra-226 and K-40.
- typical radionuclides of uranium contamination were added associated with target production

Radionuclide	ETM/DTM/Hybrid
Mo-93	DTM
Nb-93m	DTM
Tc-99	DTM
Cd-113m	DTM
Sn-119m	DTM
Cs-137	ETM
Ce-144	ETM
Pm-145	ETM
Sm-145	ETM
Pm-147	DTM
Gd-153	ETM
Tb-157	DTM
Lu-174	ETM
Hf-178n	ETM
Po-208	DTM
Po-209	DTM
Pb-210	DTM
Ac-227	DTM
Ra-228	DTM
Th-228	DTM
Pa-231	DTM
U-232	DTM
U-234	DTM
U-235	Hybrid
Pu-236	DTM
Np-237	DTM
Pu-238	DTM
U-238	ETM
Pu-239	DTM



Gamma spectrometry Analysis

- Gamma spectrometry to quantify MDA for each radionuclide
- Based on the MDA value, a radionuclide can be classified to
 - ETM MDA < 0.5 declaration threshold (criteria set by waste repository)
 - Hybrid 0.5DT<MDA < DT
 - DTM MDA > DT, or no MDA (not a gamma emitter)
- Measurement set-up specifications
 - 42 cm between the drum and the detector,
 - 1800 seconds counting time,
 - Geometry is standardized,
 - Double measurements every 20 drums (random quality controls),

				Edit di	mension	s - Simp	le Cylind	re			2
Descri	ption: Cyl-side@42c	m									OK
Comme	ent 🗍										Cancel
No.	Description	d.1	n , d.2	п d.3	d.4	d.5	Mater	ial	Density	Rel. Conc.	Apply
1	Conteneur	1.25	571.5	855			304ss	•	7.81		
2	Source - Couche	855					pvc	٣	0.2	1.00	Help
3	Source - Couche	0						٣	0	0.00	
4	Absorber 1	0						٣	0		View Drawing
5	Absorber 2	0						٣	0		
6	Source-Detector	420	0	0	0	0		Ψ			





Scaling Factors

- Analytical scaling factors
 - Obtained by a ratio between specific activity of KN and DTM/hybrid
- Experimental Scaling factors
 - Obtained by radiochemical analysis of samples
 - For BEEP-ABG
 - Experimental SFs were only included for structural material of the target, taken from SHERPA
 - For target core, only analytical scaling factors were considered
- relying exclusively on Monte Carlo and analytical calculations (for contamination with alpha)
- Agreement between analytical and experimental SFs were shown in previous studies

	6 months to 3 years									
KN	Ru-106	Bi-207	Hf-172	U-238	Na-22	Co-60				
DTM/Hybrid										
H-3	1.8	306.7	1.4		3.3	2.7				
Be-10										
C-14					3.9E-03					
Cl-36										
Ar-39										
Ca-41					9.2E-04					
V-49						3.2				
Fe-55					17.7	43.2				
Ni-63						1.1				
Zn-65	3.7E-03	0.4	3.0E-03							
Ge-68	9.1E-04	0.2	9.5E-04							
Kr-85	2.8E-02	0.6								
Sr-90	0.1	0.8								
Mo-93		1.2E-03								
Nb-93m										
Tc-99										
Ag-108m	2.7E-04	4.8E-03	5.3E-06							
Cd-113m										
Sn-119m	0.3									
Sb-125	0.3	0.1								
Pm-147	0.2									
Gd-148	2.7E-04	0.3	1.4E-02							
Eu-152										
Tb-157										
Ta-179	3.3E-02	25	3.1							
Pt-193		10								
Au-195	2.5E-02	80								
Pb-202										
TI-204		184.1								
Po-208	0.1									
Po-209										
Ph-210	7 1E-03									



Calculation of hazard factor - IRAS

Extracts from gamma-spec

- Meta data, Detected radionuclides, MDA
- Example of different criterion for different RN type Apply the criteria ۲ Radionuclide Radionuclide ETM activity contributes to IRAS if it is > calculate activities • DT(ANDRA) activity maybe reported activity or MDA Activities of ETMs ۰ Hybrid detected activity, Activities of DTMs/Hybrid • if not, calculated activity(KN_activity, SF_KN) Calculate IRAS DTM always calculated activity with SFs (analytical or ٠ experimental)
 - $IRAS = \sum_i \frac{a_i}{L_i}$,
 - a_i is the specific activity and L_i the reference activity limit for the radionuclide I





- To calculate the IRAS factor of a waste package
 - from the results of the gamma spectrometry reports and
 - from a set of scaling factors,
- To establish the list of radionuclides and their activities
- User friendly
- Possibility to add new elimination pathways and modify input data



Acknowledgement to Xavier Eric Ouvrard, the software developer.



Conclusion

• The methodology comprised several steps

- Monte-Carlo Fluka simulation
- Actiwiz calculation
- Gamma-spectrometry analysis
- Criteria application
- Establishing final radionuclide inventory, activities, IRAS.
- This methodology can be extended to other waste produced at similar facilities or research laboratories









19 September 2022



home.cern