

High-Z shielding radiopurity and material compatibility

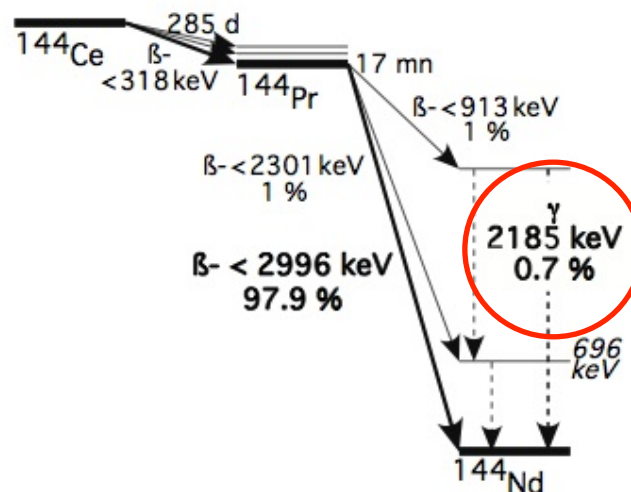
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CEA-Saclay, DSM/IRFU/SPP
CeSOX kick-off meeting
Paris, February 5th



CeANG characteristics

- Anti-neutrino emitter = ^{144}Ce - ^{144}Pr , extracted from spent nuclear fuel (SNF)
- Chemical form: Cerium oxide (CeO_2), density: $4.0 \pm 1.0 \text{ g/cm}^3$
- CeO_2 mass $\approx 10 \text{ kg}$, depending on age of SNF
- Maximum activity (@ the Mayak reprocessing plant): $140 \text{ kCi} \approx 5.2 \times 10^{15} \text{ Bq}$
- Among other γ radiation, ^{144}Ce source emits a 2.2 MeV γ -ray with 0.7% intensity. We must shield against these radiations for two reasons:
 - Biological protection during source transportation and manipulation
 - Shielding against backgrounds for a clean measurement
- We need high-Z shielding, because of the source intensity and to minimize shielding volume: tungsten is adequate



Shielding general requirements

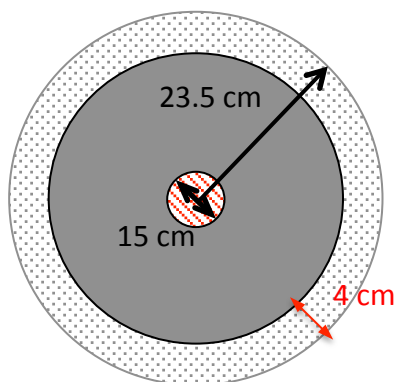
1. Biological protection, IAEA requirements (see Michel's talk):

Criteria	IAEA dose limits
Workers cat. A	≤ 20 mSv/y
Workers cat. B	≤ 6 mSv/y
Maximum dose limit per year	≤ 50 mSv

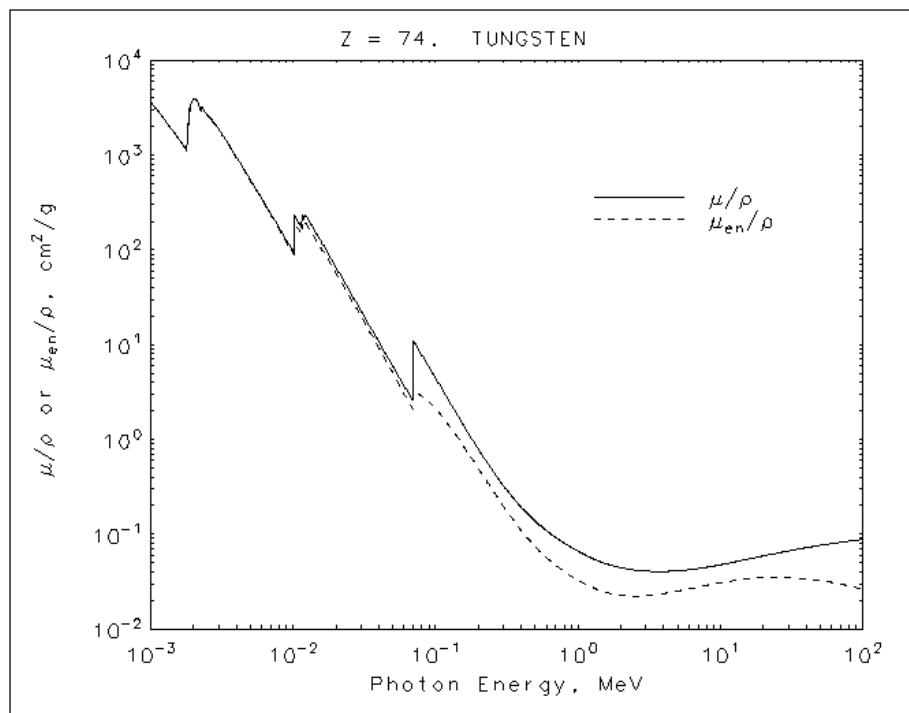
2. Background suppression requirements for our physics goal (CeLAND case):

- GEANT4 simulation on-going to precisely assess the amount of tungsten we would need to suppress the 2.2 MeV γ background to a sufficiently low level (see Vincent's talk). We need an attenuation factor of $\approx 10^{12}$ (criteria: $R_{\text{bck}} \leq 10$ Hz)
 - CeLAND phase 1: roughly 16 cm of tungsten alloy ($d=18.5$ g/cm³) along any direction (**a bit more for Borexino**)
 - CeLAND phase 2: roughly 35 cm
- GEANT4 simulations have been done to assess the maximum tolerable concentration of radionuclides (²³⁸U, ²³²Th, ⁶⁰Co, ⁴⁰K) within the shielding = radiopurity requirements (next slide)
- Background suppression requirements overcome biological protection requirements. Precise dose computations showed that a 16-cm width tungsten shielding makes easily the biological protection job.

Radiopurity GEANT4 simulations – CeLAND phase 1



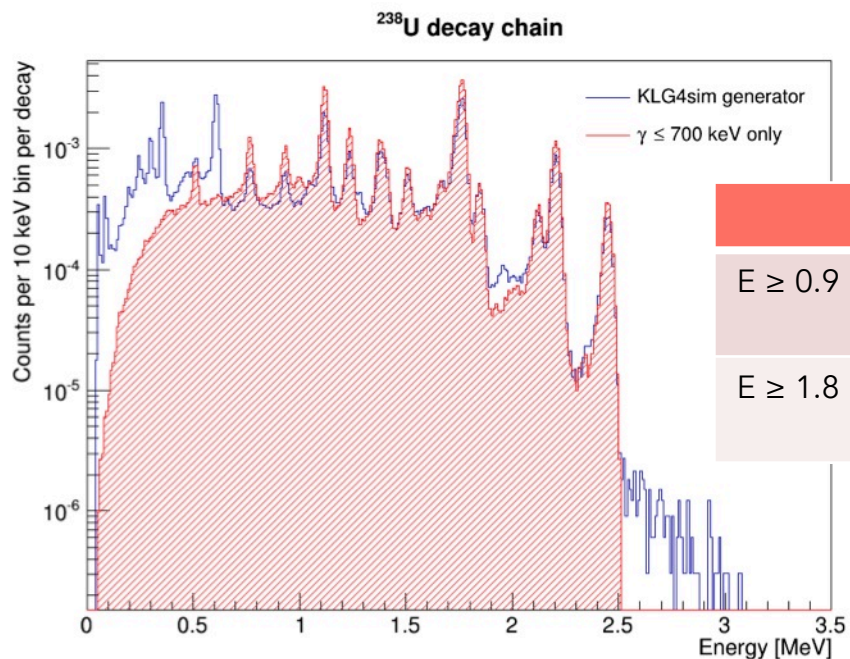
- Approximate shielding geometry to a sphere (conservative, maximizes surface over volume ratio)
- Assume a D185 like composition (97% W + 1.5% Fe + 1.5% Ni)
- Simulation filling the entire volume with γ_s is very time-consuming (high Z shielding, big attenuation)
- Simulation filling the outer part where less attenuation is expected, took a 4-cm thick outer shell, corresponds to $\geq 3.5 L_{\text{att}}$ depending on energy, i.e. we miss less than 2% of γ -rays.



$$L_{\text{att}} \approx 1.2 \text{ cm } (d_{\text{D185}} = 18.5 \text{ g/cm}^3) @ 1.5 \text{ MeV}$$

Radiopurity GEANT4 simulations – CeLAND phase 1

- Sphere filled with random γ_s from ^{40}K , ^{60}Co and ^{238}U , ^{232}Th decay chains to compute the probability of energy deposition in the KamLAND detector
- Decay chain simulation selecting the most relevant γ lines ($E \geq 700$ keV, $I_\gamma \geq 1\%$) to avoid the use of very heavy HEPevent files (which can be created via the use of the home built KamLAND generators)
- Validation of the procedure by comparing obtained energy spectra in the KamLAND detector with those obtained using KamLAND generators



Probabilities of energy deposition

	^{238}U	^{232}Th	^{40}K	^{60}Co
$E \geq 0.9$ MeV	3.7×10^{-7}	4.1×10^{-6}	$< 10^{-8}$	6.0×10^{-8}
$E \geq 1.8$ MeV	3.7×10^{-8}	2.1×10^{-6}	0	0

Probabilities expected to be roughly the same in Borexino

Radiopurity requirements for CeLAND phase 1

- Criteria: $R_{\text{bck}} \leq 5$ Hz (this criteria can't be relaxed), $d=18.5$ g/cm³, 16-cm thick spherical shielding:

Radionuclide	Max activity [Bq kg ⁻¹]	Max concentration [g/g]
²³⁸ U	32.0	$2.6 \cdot 10^{-6}$
²³² Th	3.0	$7.1 \cdot 10^{-7}$
⁶⁰ Co	≈ 200	-
⁴⁰ K	≈ 1200	$4 \cdot 10^{-2}$

- Radiopurity of tungsten shouldn't be a problem for CeLAND phase 1....
- ... even if we harden the background criteria to $R_{\text{bck}} \leq 1$ Hz or less.

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Samples provided by tungsten suppliers

- CEA-Saclay is currently in touch with 2 suppliers
 - Plansee (France)
 - Xiamen Honglu Tungsten Molybdenum Industry (China)
- We obtained several samples that were measured in Ge counters to assess radiopurity:

Alloy type	Density [g/cm ³]	²³⁸ U [mBq/kg]	²³² Th [mBq/kg]	⁶⁰ Co [mBq/kg]	⁴⁰ K [mBq/kg]
D185 plate	18.5	551 +/- 23	231 +/- 16	≤ 1.5	80 +/- 18
D185 powder	-	112 +/- 11	79 +/- 11	≤ 1.8	90 +/- 40
5% Ni-Cu	17.75-18.35	≤ 63	24 +/- 3	≤ 0.44	≤ 20
3% Ni-Cu	?	≤ 69	19 +/- 2	≤ 0.23	≤ 20
3% Ni-Fe	18.25-18.85	≤ 100	22 +/- 3	≤ 0.36	43 +/- 10

Plansee

Xiamen

- Xiamen samples much cleaner than Plansee samples
- All samples easily pass the radiopurity requirements

Material compatibility requirements

- Some questions about material compatibility have been addressed by both tungsten suppliers:
 - Tungsten porosity in water? Not a problem, tungsten alloy is manufactured by sintering at very high temperature in a liquid phase
 - Tungsten corrosion in water? Should not be a problem. Have never been encountered to the supplier knowledge.
 - 10 deg water (KamLAND outer veto water): **do we need that test for Borexino?**
 - 50 deg water (calorimeter device circulating water)
- Precise corrosion tests might be contracted to the tungsten suppliers if not too much expensive:
 - Preliminary corrosion tests have been performed by Xiamen (for free). **Results in next slides**
 - Plansee provided us with a quotation: 3 k€
- Corrosion tests will be started soon at Saclay.

Results of Xiamen corrosion test in water

- W(97%) + Fe(1.5%) + Ni(1.5%) samples (10 x 10 x 100 mm) tested in 24 and 80 deg water against corrosion, according to an experimental protocol written by Saclay.
- Exposition: 1 month
- Mass loss measurement + conductometry & pH measurement of purified water before and after exposition

Exposition in 24 deg water

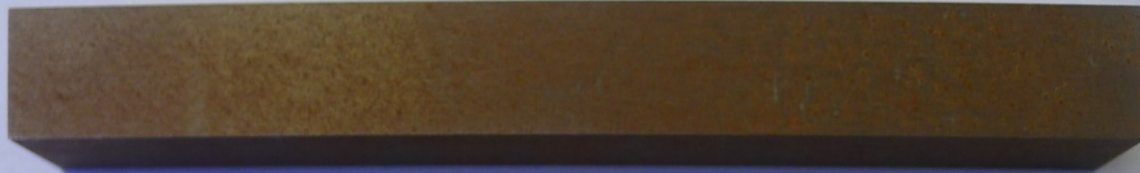


Exposition in 80 deg water



Results of Xiamen corrosion test in water

Sample I in purified water (24 °C)

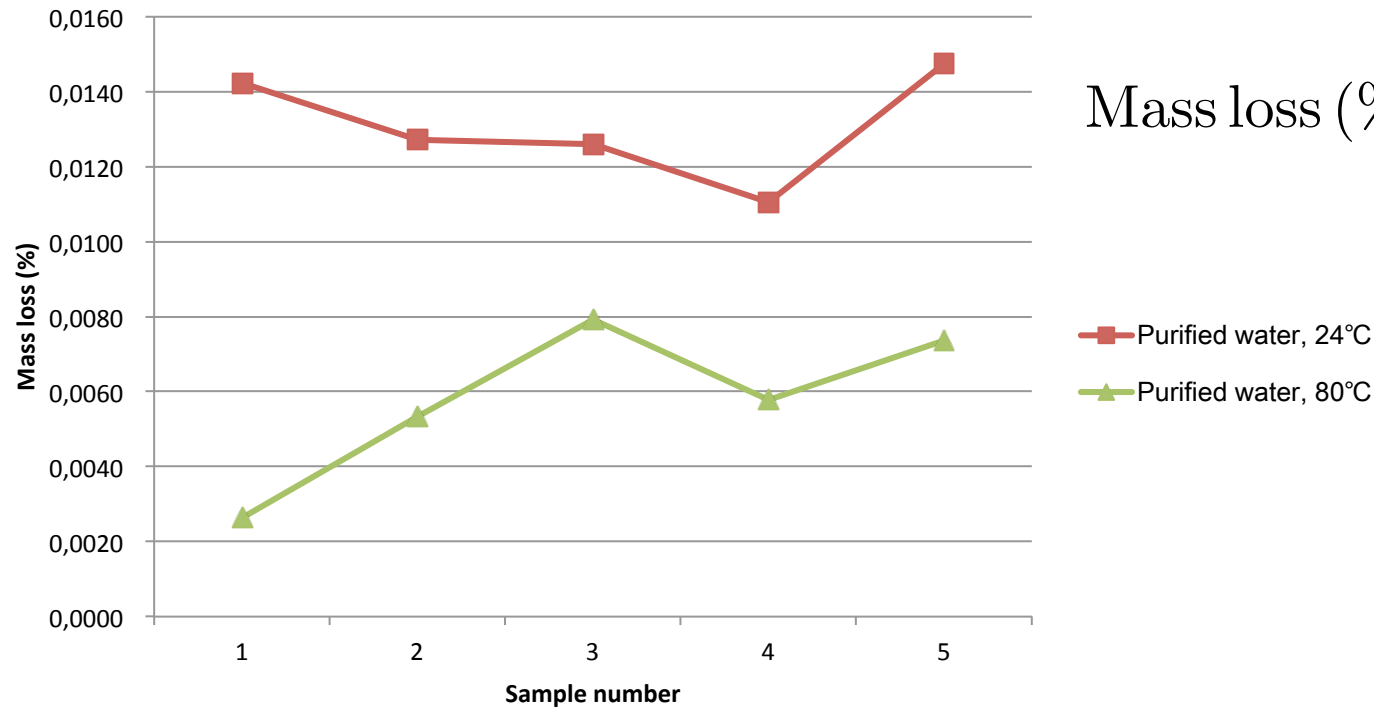


Sample II in purified water (80 °C)



Mass loss

$$\text{Mass loss (\%)} = \frac{m_i - m_f}{m_i}$$



- Extrapolated to the final tungsten shielding dimensions (cylinder of 543 mm height x 540 mm diameter) , that roughly makes a 6.2 g mass loss for a 1-month exposure. Total mass of tungsten shielding is estimated to be 2.3 tons.

Conclusion

- Radiopurity requirements are easily passed for CeSOX.
- First corrosion tests performed by Xiamen are worrisome... Need detailed investigations.
- Definitely need corrosion tests from Plansee samples
- Tungsten alloy – water compatibility is about to be addressed soon with corrosion tests at Saclay



- Will the tungsten shielding be continuously in contact with water? (i.e. continuous calorimetric measurement)
- Any other material compatibility constraints that we should be aware of for deploying in Borexino?