Development of a low neutron emission calibration source for the GERDA experiment

M.Tarka [a], L.Baudis [a], E.Bellotti [d], G.Bruno [c], C.M.Cattadori [e], R.Dressler [b], R.Eichler [b], S.Fattori [c], A.D.Ferella [a], F.Froborg [a], U.Graf [f], R.Santorelli [a], D.Schumann [b]

[a] University of Zurich, [b] Paul Scherrer Institute, [c] L'Aquila University, [d] University Milano Bicocca and INFN, [e] INFN Milano Bicocca and University, [f] Isotope Products GmbH Berlin

Abstract

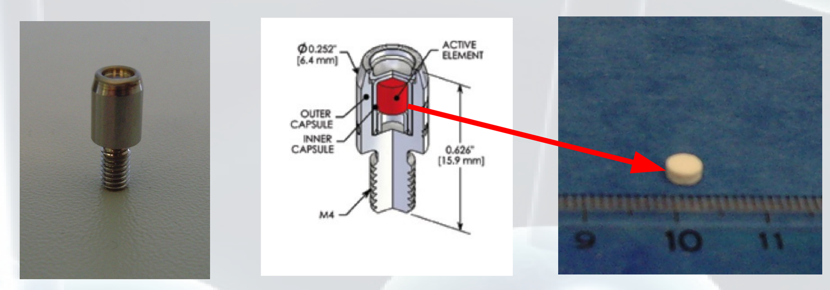
GERDA is an experiment under construction at the LNGS (Laboratori Nazionali del Gran Sasso, 3500 m.w.e.) in Italy. It will use an array of enriched ^{76}Ge detectors to search for the neutrinoless double-β decay. This requires a minimized and well understood background. To reach a sensitivity of $m_\nu < 0.13$ eV in Phase II for example, a background rate of **$B \leq 1\cdot10^{-3}$ cts/(kg·y·keV)** is required. In addition to the gamma background induced by cosmic rays and natural radioactivity, neutrons from spontaneous fission and (α-n) reactions can provide a considerable contribution. ^{228}Th has been established as a good calibration source candidate for GERDA due to its γ-emission in the region of interest around $Q_{\beta\beta}$ = 2.04 MeV. The most interesting line for energy calibration is the 2.6 MeV line and its single escape peak at 2.1 MeV. This work investigates the significance of the (α-n) neutron background coming from a ^{228}Th calibration source due to its intrinsic components under the assumption that the source will be placed permanently in the GERDA-setup. For this scenario a parking position of up to 3.5 m above the detector array during data taking is assumed. For calibration runs, the source will be moved by remote control down to the detector array. First, the neutron rate from a commercial ^{228}Th source was estimated using the software package SOURCES4mv. Subsequently an alternative source production method has been developed and tested in order to minimize the neutron yield.

Commercial ^{228}Th source

Commercially available sources for cryogenic applications consist of a porous ceramic contained inside a sealed stainless steel capsule, with the ceramic saturated with the isotopes. α particles emitted by ^{228}Th interact mainly with low-Z nuclides through (α-n) reactions resulting in a neutron flux. As the ceramic is in direct contact with the radionuclides, it is the most relevant material in terms of neutron production.

Example:
^{228}Th in a $NaAlSiO_2$ ceramic

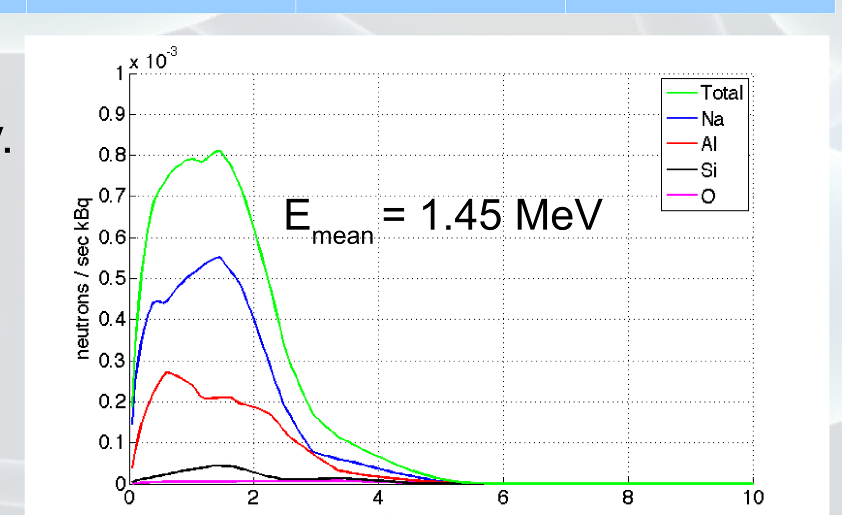
Assumptions:
- homogenously distributed
- no impurities involved


Stainless steel encapsulation — Cross section of the source — $NaAlSiO_2$ ceramic

E_α(^{228}Th-chain) = 5.2 MeV – 8.8 MeV

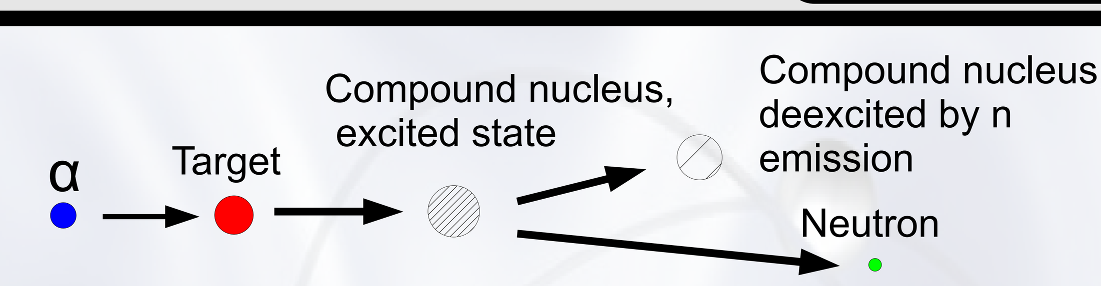
Isotope	^{23}Na	^{27}Al	^{28}Si	^{29}Si	^{30}Si	^{16}O	^{17}O	^{18}O
E_{Thr} [MeV]	3.48	3.03	9.25	1.74	3.96	15.17	< 0.1	0.85
Nat.abund. [%]	100	100	92	4.68	3.09	99.76	0.04	0.2

The neutron rates and spectra resulting from (α-n) reactions in a $NaAlSiO_2$ ceramic containing ^{228}Th were calculated with SOURCES4mv. The sum spectrum was implemented in a MC simulation in order to estimate the neutron-induced background in the energy range around $Q_{\beta\beta}$ = 2.04 MeV.



SOURCES4mv – neutron rate : $3.8\cdot10^{-2}$ n/s/kBq
MC - Resulting neutron background : $1\cdot10^{-5}$ cts/(kg·y·keV·kBq)
GERDA – total background goal : **$B \leq 1\cdot10^{-3}$ cts/(kg·y·keV)**

(α-n) reaction



$$\alpha + {}^{a}X_{b} \rightarrow {}^{a+4}Y^{*}_{b+2} \rightarrow {}^{a+3}Y_{b+2} + n$$

Reaction allowed if:
- E(α) > Coulomb barrier
- E(α) > Threshold energy E_{Thr}

$$E_{Thr} = Q(1 + m_\alpha/m_X)$$

The principle of (α-n) reactions finds its application in commercialy available neutron sources such as ^{241}Am-Be where α particles emitted by ^{241}Am produce neutrons in Be by the reaction:

$$^{4}\alpha_2 + {}^{9}Be_4 \rightarrow {}^{12}C_6 + n$$

For the GERDA experiment, such reactions must be supressed in order to minimize the neutron-induced background.

A reduction of the neutron background can be achieved by replacing the ceramic with materials exhibiting higher threshold energies for (α-n) reactions. This has been done by chemical and thermal treatment of $^{228}ThCl_4$ in a 1M HCl solution resulting in ThO_2. The process took place in a crucible made from a ~100μm thick gold foil. Gold, with a threshold energy of ~10 MeV, does not undergo (α-n) reactions in the presence of ^{228}Th α-radiation and is thus a good matrix material for the ThO_2.

Custom $^{228}ThO_2$ source

$$3ThCl_4 + 16HNO_3 \rightarrow 200°C \rightarrow 4NOCl + 8H_2O + 4Cl_2 + 3Th(NO_3)_4$$

Evaporation

750 °C

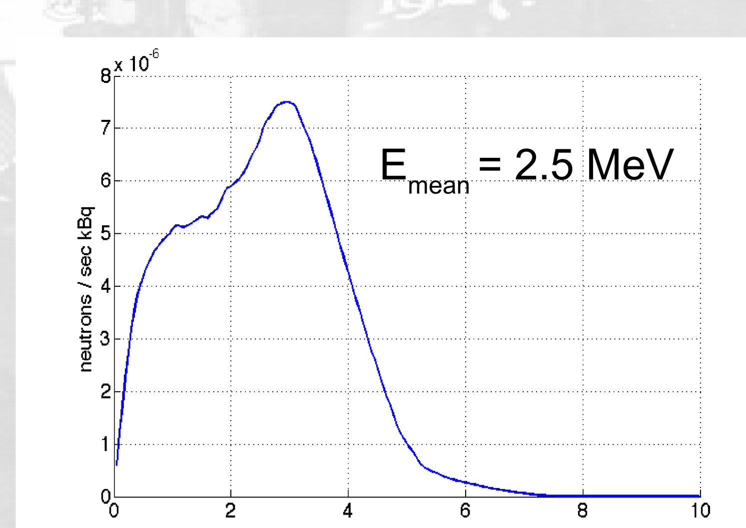
$12NO_2 + 3O_2$ + $3ThO_2$


Au - crucible

^{220}Rn gas development breaks the Th chain equilibrium

- ^{16}O: 99.76 % , E_{Th} = 15.171 MeV
- ^{17}O: 0.038 % , E_{Thr} = < 0.1 MeV
- ^{18}O: 0,205 % , E_{Thr} = 0.851 MeV
- ^{197}Au: 100 % , E_{Thr} = 9.937 MeV

Endproduct: ThO_2 in goldfoil



SOURCES4mv – neutron rate : $5\cdot10^{-4}$ n/s/kBq
MC - Resulting neutron background : $8.6\cdot10^{-8}$ cts/(kg·y·keV·kBq)
→ Expected background reduction by a factor of 116

Results

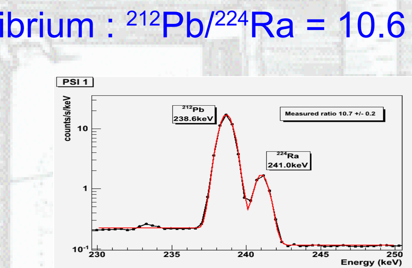
Chain restoration

$^{228}Th \rightarrow {}^{224}Ra \rightarrow {}^{220}Rn \rightarrow {}^{216}Po \rightarrow {}^{212}Pb \rightarrow {}^{212}Bi \rightarrow$ (^{212}Po / ^{208}Tl) $\rightarrow {}^{208}Pb$

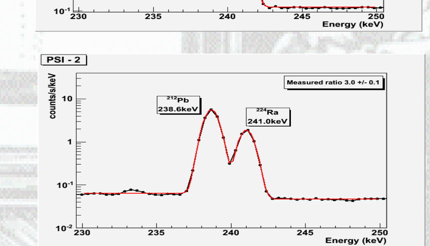
$T_{1/2}$: 1.9a, 3.7d, 55.6s, 0.15s, 10.6h, 60.5m, 2m/0.3μs

Relative peak height ratio in equilibrium : $^{212}Pb/^{224}Ra$ = 10.6

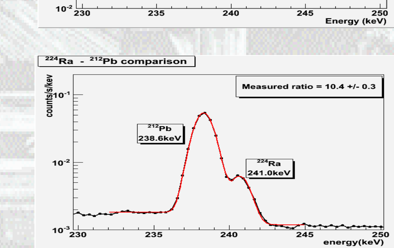
Before treatment:
$^{212}Pb/^{224}Ra$ = 10.7 ± 0.2



1h after treatment
$^{212}Pb/^{224}Ra$ = 3 ± 0.1

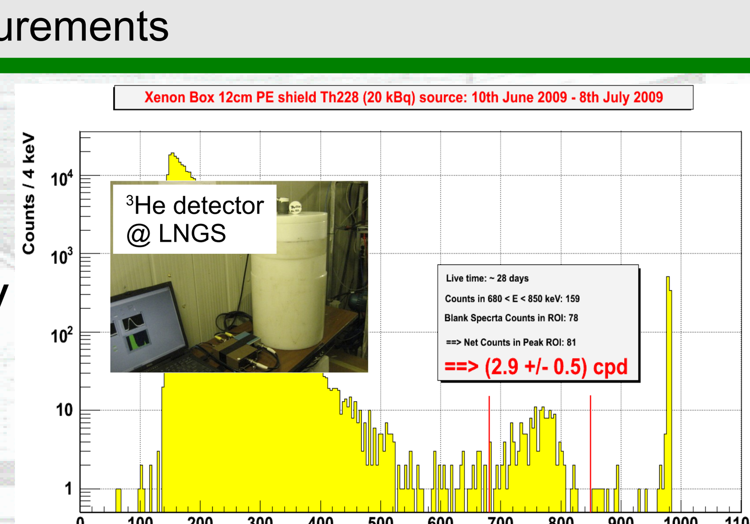


2 month after treatment
$^{212}Pb/^{224}Ra$ = 10.4 ± 0.3



20kBq ThO_2 source - Neutron measurements

The neutron rate produced by the $^{228}ThO_2$ source was measured with a ^{3}He detector located in the LNGS laboratory. The neutrons are thermalized to ~0.03 eV using 12.5 cm of PE between the source and the ^{3}He tube and counted via the energy release of 764 keV in the $^{3}He(n,p)^{3}H$ reaction.

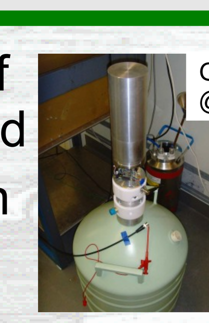


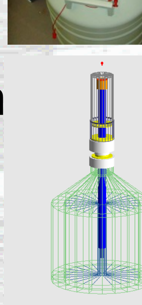
Detector efficiency:
$\varepsilon_{tot} = \varepsilon_{geom}\cdot\varepsilon_{therm}\cdot\varepsilon_{capt}$ = 0.2 %
ε_{geom} = geometrical eff.
ε_{therm} = n-thermalization eff. in PE
ε_{capt} = therm. n capturing eff. in ^{3}He

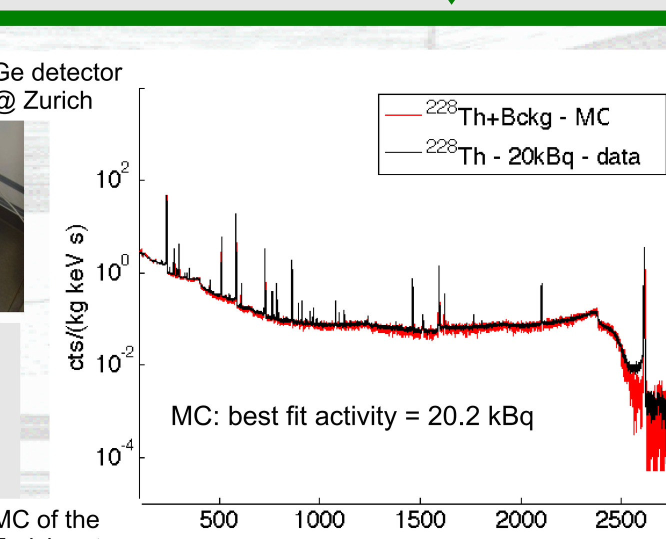
Measured n-rate: R = 8.5 ±1.5 $\cdot10^{-4}$ n/s/kBq
SOURCES4mv : R = 5 $\cdot10^{-4}$ n/s/kBq

Activity loss during chemical and thermal treatment

The activity loss during the treatment of the $^{228}ThCl_4$ solution has been estimated by comparing the ^{228}Th γ-spectra, taken with a 4x4cm Ge detector to Monte Carlo simulations. The nominal activity of the $^{228}ThCl_4$ solution was given at 20 kBq, and the best fit between data and Monte Carlo simulations resulted in an activity of 20.2 ± 0.4 kBq.


Ge detector @ Zurich


MC of the Zurich setup



→ no measured activity loss during the treatment of the $^{228}ThCl_4$ solution.