High Z Bolometers for Analysis of Internal $\beta$ and $\alpha$ Activities

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CdWO$_4$ bolometers were realized with the final purpose to study Double Beta Decay of $^{116}$Cd. The test device we developed performs as a very efficient $\gamma$ detector, with a preliminary resolution of about 5 keV FWHM. The detector background spectra are dominated at low energies by $\beta$ decay of $^{112}$Cd: the calorimetric measurement of this process allows to determine the end point $(318.8 \pm 1.4 \pm 5$ keV) and with unprecedented precision the life time $((9.3 \pm 0.5 \pm 1) \times 10^{15}$ y). Following the same approach, we have analysed the internal contamination in $^{210}$Pb of three different types of lead used for shielding in low background experiments. Various lead bolometers were developed and operated, using common modern lead, special low activity modern lead and 2000 years old Roman lead. The presence of $^{210}$Pb was clearly observed through the $\alpha$ decay of its daughter $^{210}$Po in the modern samples, while only a limit was set for the much purer Roman lead. The purifying effect of crystallization was clearly demonstrated.

1. INTRODUCTION

Using common radiation detectors, it is often difficult to measure with precision low levels of $\alpha$, $\beta$ and (at low energies) $\gamma$ activities. The reason is that in most cases it is not possible to develop a conventional detector containing the long living nuclei: therefore the source must be separated from the detector. This implies that only a small volume at the surface of the source can be examined and that, in case of $\alpha$ and $\beta$ activities, the radiation energy is degraded in the source itself. The wide choice of materials assured by bolometric technique allows to perform $\alpha$ and $\beta$ spectroscopy on the entire bulk of the sample. The consequent high efficiency and energy resolution for the process to be studied permit to determine contamination of very long living nuclei (like $^{238}$U, $^{232}$Th and $^{40}$K) with sensitivities comparable in some cases with those achievable through mass spectroscopy methods$^1$, and definitely better when lower lifetime nuclides are involved. In addition, bolometric investigation of low activities often does not imply the destruction of the material to be examined: this resource is very appreciated when the substance under
test must be employed in a low activity shielding apparatus; in this case indeed it is important to minimize the differences between the material under test and that which will be actually used for the shielding. In this paper, we report on the bolometric measurements of two unstable nuclides: $^{113}\text{Cd}$, whose $\beta$ decay is studied with a CdWO$_4$ detector, and $^{210}\text{Po}$, which derives from $^{210}\text{Pb}$ (often present in common lead) and undergoes an $\alpha$ disintegration, clearly observable in lead bolometers.

2. $^{113}\text{Cd} \beta$ SPECTRUM MEASURED WITH A CdWO$_4$ BOLOMETER

We have planned to develop detectors containing Cd because $^{116}\text{Cd}$ is an interesting candidate for neutrinoless Double Beta Decay (DBD), a never detected nuclear process which is a unique probe to test lepton number non conservation and neutrino properties. The DBD transition energy of $^{116}\text{Cd}$ is reasonably high (2802 keV), implying large phase space for the decay and low background; in addition, $^{116}\text{Cd}$ isotopic abundance is not desperately low (7.49%). A non negligible drawback is represented by the high neutron cross section for (n,$\gamma$) reactions, whose negative effects on background should however be substantially reduced by an underground operation of the detector.

Among Cd compound, CdWO$_4$ is rather appealing for three reasons: firstly, its Debye temperature, that we did not succeed to find in literature, should be rather high, as suggested by the hardness and the high fusion temperature of this compound; secondly, large single crystals are commerciably available; finally, CdWO$_4$ is an excellent scintillator, opening the way to a simultaneous measurement of heat and light which could suppress the $\alpha$ background, which is very critical around 3 MeV, just where we expect the DBD counts.

We have developed a preliminary detector consisting of a CdWO$_4$ single crystal, with cylindrical shape (1.5 cm high and 2.5 cm diameter) and a mass of 58 g. The detector was mounted in a copper box, where it is held by 8 (3 below, 1 above and 4 around) spring loaded tips. A copper wire, glued with GE varnish, constitutes the thermal link to the heat bath. The temperature sensor is a NTD Ge thermistor provided us by E.E. Haller, with a volume of roughly 4x3x1 mm$^3$.

Fig. 1 Calibration spectrum of CdWO$_4$ bolometer: seven peaks from the source can be identified.
A small silicon chip (1x1x0.5 mm$^3$) carrying a metal film 10 MΩ resistor was also glued to the crystal for stabilization purposes (calibrated square voltage pulses can be injected in the resistor in order to deliver fixed amounts of energy to the crystal).

After preliminary tests in our dilution refrigerator in Milan, where it was not possible to appreciate fully the detector potentialities because of cosmic ray pile-up, the detector was moved to Gran Sasso Laboratories and mounted in our second Oxford Instruments dilution refrigerator, built according to the same criteria as the first one, where a DBD experiment on $^{130}$Te is running.$^1$

The detector base temperature was 19.5 mK. Through a series of 150 metal film resistors (for a total resistance of 1.5 GΩ) thermally anchored at the mixing chamber, 7.7 mV bias was applied at the thermistor, corresponding to a resistance and temperature of 4.4 MΩ and 25.3 mK respectively.

When exposed to $\gamma$ calibration sources through a window in the lead and copper external shields, spectra largely deteriorated by long term gain fluctuations (due probably to temperature instabilities having the same trend as the helium level in the main bath) were obtained. The calibrated pulses through the 10 MΩ resistor allowed to stabilize the spectra: the resolution at low energies was then about 5 keV FWHM (fig. 1). A residual linear dependance of the resolution on the energy can be attributed to a non complete stabilization: in fact, the thermal pulses generated by the calibration resistor have not the same shape as the particle ones (the rise time is much longer), and even the amplitude dependance on time is different. This discrepancy does not allow a full stabilization of the spectra. An alternative method to inject known amounts of energy in the crystal consists of light pulses directed through an optical fiber to a black spot on the crystal itself. This technique was already tested in Milan with a large LiF bolometer, with very encouraging results. Pulse heights, in a naive calorimetric approach, correspond to a heat capacity of 1.2x10$^{-9}$ J/K.

A stabilized background spectrum of about 158 h of effective running time was collected. As expected, the low energy branch of the spectrum is dominated by $\beta$ decay of $^{113}$Cd (fig. 2), which has an isotopic abundance of 12.2 %. A fit to the data was carried out (fig. 2) assuming a spectrum shape factor $C$ corresponding to a fourth forbidden non unique $\beta$ transition.$^5$ $C$ is given by:

$$C = p^6 + 7A_1 p^4 q^2 + 7A_2 p^2 q^4 + A_3 q^6$$
where $p$ and $q$ are the momenta of the emitted electron and neutrino respectively. The background in the $\beta$ decay region was extrapolated from higher energies assuming a shape given by a constant plus an exponential term.

The coefficients which appear in $C$ were determined to be:

$$A_1 = 0.765 \pm 0.095; \quad A_2 = 0.589 \pm 0.177; \quad A_3 = 2.04 \pm 0.74.$$ 

corresponding to the following decay parameters:

$$\tau_{1/2}^{(113)Cd} = (9.3 \pm 0.5\text{(stat.)} \pm 1\text{(syst.)}) \times 10^{15} \text{ y}$$
$$E_0^{(113)Cd} = 318 \pm 1.4\text{(stat.)} \pm 5\text{(syst.)} \text{ keV}$$

The source of the systematic errors are the uncertainties in the evaluation of the shape factor and the procedure of background extrapolation.

### 3. INTERNAL $\alpha$ ACTIVITIES IN LEAD BOLOMETERS

Due to high atomic number and density, acceptable mechanical properties and reasonable cost, lead is probably the most important shielding material in low background experiment. It is therefore crucial to determine intrinsic lead radioactivity. The main contaminant in commercial lead is normally $^{210}$Pb and its daughters $^{210}$Bi and $^{210}$Po. The relatively long lifetime of $^{210}$Pb (22.3 y) often makes this nuclide much more abundant than expected from secular equilibrium, with the exception of old lead. $^{210}$Pb emits very soft $\beta$ and $\gamma$ rays, usually self-absorbed. On the contrary its daughter $^{210}$Bi emits energetic electrons ($E_{\text{max}} = 1.16 \text{ MeV}$), that can be only indirectly detected through bremsstrahlung and Pb characteristic $x$-rays. $^{210}$Po, daughter of $^{210}$Bi, decays mainly to the stable $^{206}$Pb emitting a $5.304 \text{ MeV} \alpha$ particle. This contamination prevents usually from facing directly modern lead samples to the detectors to be shielded. To study $^{210}$Pb contamination through $^{210}$Po $\alpha$ decay, we have developed and operated various lead bolometers. We report here on the results obtained with 4 samples, made respectively with polycrystalline modem lead (PML), crystalline modern lead (CML), special low activity modem lead (JML) and 2000 year old Roman lead (RL). Except CML, that was purposely crystallized, the other samples were simply cut or melted from the respective ingots.

Lead is a superconductor. This poses in principle some problems about its utilization as an energy absorber in a phonon mediated detector, since the quasiparticle yield might dominate the phonon yield at low temperatures. There are however some encouraging results about this topic. Narayananmurti et al.\textsuperscript{6} has measured the quasiparticle number decay time $\tau_{\text{eff}}$ in lead as a function of the temperature: the results show that above 1.8 K $\tau_{\text{eff}}$ increases exponentially as the temperature decreases (as expected from the theory), but below this value $\tau_{\text{eff}}$ tends to stabilize; its value is $0.5 \mu$s at 1.25 K and seems to saturate. Lower temperature data are not available, but typical bolometric integration times are at least 3 orders of magnitude longer than the $\tau_{\text{eff}}$ value at 1.25 K, suggesting that complete conversion of energy into phonons on a bolometric time scale should be possible even below 0.1 K, at which we have operated our devices. Furthermore, a more general approach followed by N.E. Booth et al.\textsuperscript{7} shows that in
superconductors with high critical and low Debye temperatures (like lead, for which $T_c = 7.19$ K and $\Theta_D = 82$ K) the phonon yield should quickly prevail on quasiparticle yield. Other authors\(^9\) have found that in superconductors the thermalization efficiency of the energy deposited by a particle drops abruptly when $T < 3 \times 10^{-4} \Theta_D$, corresponding to 25 mK in lead. We have operated most of the bolometers\(^9\) around or above this temperature, obtaining signals in reasonable agreement with what expected form the theoretical heat capacity. In one case (JML) we have worked at 16.4 mK: at that temperature indeed the signal was about 20 times lower than expected, confirming this threshold effect.

Several types of detector mountings and heat-sinkings were tested. Better results were obtained holding the detector, cut in a rectangular shape, with 10 spring loaded tips, distributed in equal number on two opposite sides. A NTD Ge sensor (roughly $4 \times 1 \times 0.3$ mm\(^3\)) was glued with GE varnish to each crystal. Thin mylar layers ($23 \mu m$ thick) were interposed both between sensor and absorber and between tips and absorber, to provide electrical decoupling.

First tests without a careful insulation between thermistor and lead exhibited pulse shapes characterized by a fast initial component (integrated by our parasitic capacitance), which disappeared completely after mylar interposition. A price to pay for this operation was of course a very long rise time (between 10 and 20 ms) which surely implied a remarkable reduction of the signal amplitude. The operation parameters of the 4 mentioned samples are reported in table I. As bolometric performances are concerned, no difference was observed between crystalline and polycrystalline samples. A typical background energy spectrum, obtained with JML, is reported in fig. 3: the $\gamma$ peaks at 1461 and 2615 keV (due respectively to $^{40}$K and $^{208}$Tl) can be clearly appreciated, while at high energy a small peak (4 % FWHM) centered at 5.4 MeV indicates the presence of $^{210}$Po in the sample.

The $^{210}$Po activities for the various samples are reported in table I. One observes that the high contamination present in PML (about 100 Bq/Kg) can be reduced by one order of magnitude by crystallizing the material, as the residual activity of CML shows. A small contamination is still present in JML, provided us by the company Johnson&Matthey, while no sign of $^{210}$Po was found in RL. Roman lead is therefore the best candidate for very low activity shieldings\(^9\).
4. CONCLUSIONS

We have shown in two different cases that bolometric technique, thanks to its unique flexibility in the choice of detector materials joined to the accurate energy spectroscopy that it provides, is a powerful method to determine the parameters of intrinsic internal radioactivities. In particular, we have fully studied the activities coming from $^{113}$Cd in natural Cadmium and from $^{210}$Po (originated by $^{210}$Pb) in several types of lead.

REFERENCES

2. The use of the bolometric technique to investigate radioactive contaminations in Pb was suggested to us by H.H. Andersen