# EXPERIMENTAL INVESTIGATION OF <sup>106</sup>Cd DOUBLE BETA DECAY USING AN ENRICHED <sup>106</sup>CdWO<sub>4</sub> SCINTILLATOR

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This article describes an experiment to search for the double beta decay of <sup>106</sup>Cd using an enriched <sup>106</sup>CdWO<sub>4</sub> scintillation detector in coincidence with two CdWO<sub>4</sub> scintillation counters at the Gran Sasso underground laboratory of the National Institute for Nuclear Physics (LNGS INFN, Italy). The obtained limits on different modes and channels of the process are among the strongest in the field.

#### **INTRODUCTION**

Double beta decay is one of the rarest nuclear processes, with the half-lives ranging from  $T_{1/2}=10^{18}$  to  $10^{24}$  years [1, 2]. The process with increase of the nuclear charge by two units, accompanied by the emission of two electrons and two anti-neutrinos  $(2\nu 2\beta^{-})$ , has been observed in 11 nuclei. Additionally, "double beta plus" processes, in which the nuclear charge decreases by two units, are also possible for more than thirty nuclei. These include double electron capture (2EC), electron capture accompanied by positron emission (EC $\beta^+$ ), and double positron decay  $(2\beta^{+})$ . However, experimental sensitivity to these double beta plus processes is significantly lower. There are three indications of 2v2EC decay in <sup>78</sup>Kr [3], <sup>130</sup>Ba [4, 5]), and <sup>124</sup>Xe [6–8].



Fig. 1. A simplified  $2\beta$  decay scheme of <sup>106</sup>Cd

The <sup>106</sup>Cd nuclide is a promising candidate for the search for double beta plus decays due to several advantageous properties. It has a relatively high transition energy of  $Q_{2\beta}=2775.39(10)$  keV [9] and a notable isotopic abundance of  $\delta$ =1.245(22) % [10]. A simplified decay scheme of <sup>106</sup>Cd is shown in Fig. 1. Additionally, enrichment can be achieved using gas centrifugation methods. An essential factor is the availability of well-established cadmium purification techniques and the technology for producing highquality radiopure cadmium tungstate (CdWO<sub>4</sub>) crystal scintillators [11, 12], which enable calorimetric experiments with high detection efficiency. This approach was utilized in the current experiment aimed at searching for the  $2\beta$  decay of <sup>106</sup>Cd.

## **EXPERIMENTAL SETUP**

The experiment is conducted at a depth equivalent to 3.8 km of water in the Gran Sasso underground laboratory of the National Institute for Nuclear Physics (LNGS INFN, Italy). The main part of the detector system consists of three CdWO<sub>4</sub> crystal scintillators. The central scintillator, nearly cylindrical in shape, measures Ø 27  $\times$  50 mm, has a mass of 215.4 g, and is enriched to 66% with <sup>106</sup>Cd (<sup>106</sup>CdWO<sub>4</sub>) [12]. The scintillator is viewed by a low-radioactivity Hamamatsu R11065-MOD photomultiplier tube (PMT) through a high-purity quartz light guide ( $\emptyset$  66  $\times$  100 mm) and a polystyrene-based plastic scintillator (Ø 40mm × 83 mm).

Two large-volume CdWO<sub>4</sub> scintillators (Ø 70  $\times$ 38 mm), containing cadmium with its natural isotopic abundance, feature cylindrical cutouts to closely enclose the <sup>106</sup>CdWO<sub>4</sub> crystal. These scintillators are viewed by radiopure Hamamatsu R6233MOD PMTs via highpurity quartz light guides (Ø 70 × 200 mm). Opticalgrade silicone grease ensures proper optical contact between the detector components, while Teflon elements hold the assembly together to maintain the detector system's geometry. To minimize background caused by radioactive contamination from the PMTs, the detectors are shielded with high-purity copper bricks ("internal copper"). The entire system is housed within an 11-cm-thick high-purity copper box ("external copper"), further enclosed by layers of low-radioactivity lead (15 cm), cadmium (2 mm), and polyethylene (10 cm) for additional background suppression. A schematic diagram of the experimental setup is shown in Fig. 2.



Fig. 2. Schematics of the experimental set-up. PMT is photomultiplier tube. OC is for optical couplant Dimensions are given in mm

## **PULSE-SHAPE DISCRIMINATION**

The difference in pulse shapes for  $\gamma \& \beta$  and  $\alpha$  scintillation signals was utilized to suppress background contributions from  $\alpha$  particles. A numerical parameter, so called Shape Indicator (*SI*), was calculated for each signal. Further details on pulse-shape discrimination of scintillation signals using the optimal filter method can be found italic [13]. The dependence of *SI* on energy for one of the CdWO<sub>4</sub> detectors is presented in Fig. 3, demonstrating a clear discrimination between signals of  $\alpha$  and  $\gamma \& \beta$  particles.

It is important to note that the energy of  $\alpha$  particles  $(E_{\alpha})$  associated with the U/Th decay chains ranges from 4.0 to 8.8 MeV. However, due to the quenching of scintillation light yield for  $\alpha$  particles in the  $\gamma$  energy scale, the  $\alpha$  spectrum appears shifted to lower energies [14]. The  $\alpha/\gamma$  ratio (i.e., the ratio of the energy of an  $\alpha$  particle in the  $\gamma$  scale to its actual energy) was measured for the <sup>106</sup>CdWO<sub>4</sub> detector as  $\alpha/\gamma=0.12(2)+0.011(2)\times E_{\alpha}$ , and for the CdWO<sub>4</sub> counters as

 $\alpha/\gamma$ =0.08(1)+0.015(2)× $E_{\alpha}$ , where  $E_{\alpha}$  is in MeV [15]. The analysis of  $\alpha$  spectra allowed for the determination of from the U/Th decay chains [15].crystal contamination by  $\alpha$ -active daughter nuclides from the U/Th decay chains [15].



Fig. 3. Two-dimensional distribution of background events measured by the CdWO<sub>4</sub> detector, showing energy (E) versus Shape Indicator (SI) coordinates (top). The projected energy spectra of separated  $\alpha$ and  $\gamma \& \beta$  events (bottom)

#### EXPERIMENTAL ENERGY SPECTRA

The experimental energy spectra recorded over 1075 days by the <sup>106</sup>CdWO<sub>4</sub> and CdWO<sub>4</sub> detectors under various selection conditions are shown in Fig. 4. The dominant component of the spectra measured by the <sup>106</sup>CdWO<sub>4</sub> detector below 0.7 MeV, both in anticoincidence (AC) and in coincidence (CC) with the CdWO<sub>4</sub> counters, corresponds to the  $\beta$ -decay of <sup>113m</sup>Cd, which is present in the <sup>106</sup>CdWO<sub>4</sub> crystal [12, 16]. Similarly, the  $\beta$  spectrum of <sup>113m</sup>Cd constitutes the main contribution to the spectra measured by the CdWO<sub>4</sub> detectors below 0.6 MeV.

An important selection condition is coincidences with a 511 keV  $\gamma$  quantum in the CdWO<sub>4</sub> counter(s), which occur after annihilation of a positron emitted in double beta processes in <sup>106</sup>Cd. This condition is illustrated in Fig. 4 ( $\gamma$ & $\beta$ -CC,  $E^{nat}$ =511±2 $\sigma_E$  keV, where  $E_{nat}$  is the energy deposition in at least one of the CdWO<sub>4</sub> detectors,  $\sigma_E$  is energy resolution of the CdWO<sub>4</sub> detectors).

#### RADIOACTIVE CONTAMINATION OF THE EXPERIMENTAL SETUP

To interpret the experimental data collected by the  ${}^{106}$ CdWO<sub>4</sub> detector above 0.7 MeV in anti-coincidence (AC) mode and above 0.4 MeV in coincidence (CC) mode, a background model was developed, incorporating the following components:

1. Contributions from  ${}^{40}$ K,  ${}^{232}$ Th, and  ${}^{238}$ U (including their decay chains);

2. Residual  $\alpha$  events distribution in the  $^{106}CdWO_4$  crystal;

3.  $\beta$  decay of <sup>176</sup>Lu and <sup>113m</sup>Cd, as well as  $2\nu 2\beta$  decay of <sup>116</sup>Cd with a half-life of  $T_{1/2}=2.63\times 10^{19}$  years in the <sup>106</sup>CdWO<sub>4</sub> scintillator;

4. <sup>113</sup>Cd contamination in both the CdWO<sub>4</sub> and  $^{106}$ CdWO<sub>4</sub> crystal scintillators;

5. <sup>56</sup>Co and <sup>60</sup>Co in the internal copper shielding.



Fig. 4. Energy spectra measured by <sup>106</sup>CdWO<sub>4</sub> (top) and both CdWO<sub>4</sub> detectors (bottom) for 1075 days under the different selection conditions.  $\gamma \& \beta$  denotes spectrum of  $\gamma \& \beta$  events. AC (CC) means anticoincidence (coincidence) mode.  $E^{nat} = 511 \pm 2\sigma_E \text{ keV}$ denotes energy deposit in at least one of the CdWO<sub>4</sub> detectors in the energy interval  $511 \pm 2\sigma_E \text{ keV}$ , where  $\sigma_E$ is the standard deviation of energy resolution.  $E^{106} >$ 500 keV denotes energy deposit in the <sup>106</sup>CdWO<sub>4</sub> detector more than 500 keV

The energy distributions of these background sources were simulated using the Monte Carlo package EGSnrc [17], with the initial kinematic conditions provided by the DECAY0 event generator [18]. Fig. 5 presents the results of a simultaneous binned maximum likelihood fit of five  $\gamma \& \beta$ -spectra: three spectra recorded by the <sup>106</sup>CdWO<sub>4</sub> detector under AC, CC, and CC  $E^{\text{nat}}=511\pm 2\sigma_{\text{E}}$  keV conditions, as well as two spectra measured by the CdWO<sub>4</sub> detectors under CC and CC  $E^{106}$ >500 keV conditions. The estimated radioactive contaminations of the experimental setup components were derived from this fitting procedure. Upper limits of radioactive contaminations by <sup>40</sup>K and U/Th families of plastic scintillator, Teflon tape and details, quartz lightguides, internal and external copper are at the level of ~1...10 mBq/kg. For optical couplant upper limits of radioactive contaminations by U/Th families are at the level of ~10 mBq/kg and by  $^{40}$ K at level of ~0.3 Bq/kg. For PMTs upper limits of radioactive contaminations by <sup>40</sup>K and U/Th families are at the level of ~0.5...2.5 Bq/kg.

## **EXPERIMENTAL SENSITIVITY**

There were no effects observed in the experimental data that could be attributed to the  $2\beta$  decay of <sup>106</sup>Cd.

Therefore, half-life limits for various channels and modes of  $2\beta$  decay were established using the formula  $\lim T_{1/2} = N(^{106}\text{Cd}) \times \ln 2 \times t/ \lim N_{dec}$ , where  $N(^{106}\text{Cd}) = 2.42 \times 10^{23}$  is the number of  $^{106}\text{Cd}$  nuclei in  $^{106}\text{CdWO}_4$ , *t* is the measurement time, and  $\lim N_{dec}$  is a maximal number of decays of the process searched for that can be excluded according to Feldman and Cousins procedure [19] with a given confidence level (C.L.). Values of  $\lim N_{dec}$  were obtained from the fit of the energy spectra with a background model plus an effect searched for. The example of experimental data fit with the background model plus 0v2EC in  $^{106}Cd$  to the ground state of  $^{106}Pd$  is shown in Fig. 5. For the different channels and modes of  $^{106}Cd 2\beta$  decay the value lim  $N_{dec}\approx 20...2200$  which gives the experimental sensitivity at the level of  $\lim T_{1/2} = 10^{20}-10^{22}$  years.



Fig. 5. Results of the combined fit of the  $\gamma$ & $\beta$  spectra easured by the <sup>106</sup>CdWO<sub>4</sub> and two CdWO<sub>4</sub> detectors unde different selection conditions. The contributions from the contamination of the setup components are shown separately (see legend)

#### CONCLUSIONS

The most sensitive experiment to search for double beta plus decay of <sup>106</sup>Cd was conducted at the Gran Sasso underground laboratory of INFN (Italy) using an enriched <sup>106</sup>CdWO<sub>4</sub> scintillator in coincidence with two large-volume CdWO<sub>4</sub> detectors. The background model of the experimental setup has been constructed and radioactive contamination of the setup materials has been estimated. After 1075 days of data taking no effects were observed. The experimental sensitivity established for different channels and modes of <sup>106</sup>Cd 2 $\beta$  decay, reaches the level of lim $T_{1/2}=10^{20}-10^{22}$  years.

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