Radioactive contamination of scintillators

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Abstract

Low counting experiments (search for double $\beta$ decay and dark matter particles, measurements of neutrino fluxes from different sources, search for hypothetical nuclear and subnuclear processes, low background $\alpha$, $\beta$, $\gamma$ spectrometry) require extremely low background of a detector. Scintillators are widely used to search for rare events both as conventional scintillation detectors and as cryogenic scintillating bolometers. Radioactive contamination of a scintillation material plays a key role to reach low level of background. Origin and nature of radioactive contamination of scintillators, experimental methods and results are reviewed. A programme to develop radiopure crystal scintillators for low counting experiments is discussed briefly.

Keywords: Scintillation detector; Rare decays; Low counting experiment; Radioactive contamination

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1 Introduction

Search for neutrinoless double beta decay, dark matter particles, measurements of solar and reactor neutrino fluxes, tests of fundamental laws with increasing accuracy (e.g. Pauli exclusion principle, charge conservation, stability of nucleons, etc.), search for hypothetical particles and effects beyond the Standard Model (axions; charge, magnetic momentum, mass of neutrino; etc.) are the topics of astroparticle physics. Scintillation detectors possess a range of important properties for low counting experiments: presence of certain chemical elements (important in searches for double decay, investigations of rare $\beta$ and $\alpha$ decays) or variety of elements (which can be exploited in dark matter detectors to probe new areas of parameter space), large sensitive volume, reasonable energy resolution (very high in a case of cryogenic scintillating bolometers), low energy threshold, long time stability of operation, pulse-shape discrimination ability, low cost.

Radiopurity is a crucial property of scintillation material to reach low background counting rate of a detector. We review here application of scintillation materials in astroparticle physics, experimental methods to measure activities of radionuclides inside scintillators, origin and nature of radioactive contamination, radiopurity data for different scintillation materials. A programme to develop radiopure crystal scintillators for low counting experiments is discussed briefly. We do not consider here noble gas based low background scintillation detectors referring readers to the reviews \cite{1,2,3}. 

1
2 Scintillators in astroparticle physics

2.1 Double $\beta$ decay

Observations of neutrino oscillations give clear evidence that neutrino is a massive particle (see e.g. [4, 5, 6, 7]). The neutrinoless double beta ($0\nu 2\beta$) decay violates the lepton number conservation and is possible if neutrino is a Majorana particle (identical to its antiparticle) with non-zero mass [8]. The process is one of the ways to investigate properties of neutrino and weak interaction, and to test the Standard Model of particle physics [9, 10, 11, 12, 13, 14]. In particular, investigation of this phenomenon could allow to determine the absolute scale of the neutrino mass and the neutrino mass hierarchy. The $0\nu 2\beta$ decay is still not observed despite the seventy years of searches (for the status of $0\nu 2\beta$ decay searches we refer reader to the reviews [15, 16, 17, 18, 19, 20] and recent experiments [22, 24, 23, 21, 25]).

High sensitivity experiments to search for double $\beta$ processes in different nuclei are strongly required both for theoretical and experimental reasons. Despite valuable theoretical efforts, there is a substantial difference in the $0\nu 2\beta$ nuclear matrix elements (NME) calculated by using different nuclear models [26]. Therefore, the theory does not provide suggestions of a nucleus with the highest decay probability. From the experimental point of view, since $0\nu 2\beta$ decay is expected to be an extremely rare process, detection of the decay in one nucleus naturally will call for support of the observation with other nuclei. Furthermore, development of experimental methods for different nuclei is required taking into account possible breakthroughs in experimental techniques [27]. The recent progress in low-temperature scintillating bolometers is a good example.

There are quite a big number of scintillation materials which contain double $\beta$ active isotopes. For this reason scintillators are widely used in double $\beta$ decay experiments. It is worth to mention a pioneer work of der Mateosian and Goldhaber to search for neutrinoless double $\beta$ decay of $^{48}\text{Ca}$ by using enriched and depleted in $^{48}\text{Ca}$ calcium fluoride [CaF$_2$(Eu)] crystal scintillators [28]. During the last two decades several high sensitivity studies of the double $\beta$ decay processes were performed using crystal scintillators with specific candidate nuclei. The most sensitive $2\beta$ experiments with crystal scintillators are reported in Table [1].
Table 1: The most sensitive calorimetric $2\beta$ experiments with scintillators. Half-life limits are given at 90% confidence level.

| $2\beta$ transition | Scintillator | Main results: half-life (channels) | Reference |
|---------------------|--------------|-----------------------------------|-----------|
| $^{40}\text{Ca}\rightarrow^{40}\text{Ar}$ | $\text{CaWO}_4$ | $\geq 9.9 \times 10^{21}$ yr ($2\nu 2\kappa$) | 29 |
|                     | $\text{CaWO}_4$ | $\geq 1.4 \times 10^{22}$ yr ($0\nu 2\varepsilon$) | 29 |
| $^{46}\text{Ca}\rightarrow^{46}\text{Ti}$ | $\text{CaF}_2(\text{Eu})$ | $\geq 1.0 \times 10^{17}$ yr ($0\nu 2\beta^-$) | 30 |
| $^{48}\text{Ca}\rightarrow^{48}\text{Ti}$ | $\text{CaF}_2(\text{Eu})$ | $\geq 5.8 \times 10^{24}$ yr ($0\nu 2\beta^-$) | 31 |
| $^{70}\text{Zn}\rightarrow^{70}\text{Ge}$ | $\text{ZnWO}_4$ | $\geq 3.8 \times 10^{18}$ yr ($2\nu 2\beta^-$) | 32 |
|                     | $\text{ZnWO}_4$ | $\geq 3.2 \times 10^{19}$ yr ($0\nu 2\beta^-$) | 32 |
| $^{64}\text{Zn}\rightarrow^{64}\text{Ni}$ | $\text{ZnWO}_4$ | $\geq 1.1 \times 10^{19}$ yr ($2\nu 2\kappa$) | 32 |
|                     | $\text{ZnWO}_4$ | $\geq 9.4 \times 10^{20}$ yr ($2\nu 2\beta^+$) | 32 |
| $^{82}\text{Se}\rightarrow^{82}\text{Kr}$ | $\text{Zn}^{82}\text{Se}$ | $\geq 2.4 \times 10^{24}$ yr ($0\nu 2\beta^-$) | 33 |
| $^{100}\text{Mo}\rightarrow^{100}\text{Ru}$ | $\text{ZnMoO}_4$ | $= 7.15 \pm 0.37(\text{stat.}) \pm 0.66(\text{syst.}) \times 10^{18}$ yr ($2\nu 2\beta^-$) | 34 |
|                     | $\text{Li}_2^{100}\text{MoO}_4$ | $= 6.90 \pm 0.15(\text{stat.}) \pm 0.37(\text{syst.}) \times 10^{18}$ yr ($2\nu 2\beta^-$) | 35 |
|                     | $\text{Li}_2^{100}\text{MoO}_4$ | $\geq 7.0 \times 10^{22}$ yr ($0\nu 2\beta^-$) | 36 |
|                     | $4\text{sthep} \text{Ca}^{100}\text{MoO}_4$ | $\geq 4.0 \times 10^{21}$ yr ($0\nu 2\beta^-$) | 37 |
| $^{106}\text{Cd}\rightarrow^{106}\text{Pd}$ | $^{106}\text{CdWO}_4$ | $\geq 1.1 \times 10^{21}$ yr ($2\nu \beta^+$) | 38 |
|                     | $^{106}\text{CdWO}_4$ | $\geq 2.2 \times 10^{21}$ yr ($0\nu \beta^+$) | 39 |
| $^{108}\text{Cd}\rightarrow^{108}\text{Pd}$ | $\text{CdWO}_4$ | $\geq 1.1 \times 10^{18}$ yr ($2\nu 2\kappa$) | 40 |
|                     | $\text{CdWO}_4$ | $\geq 1.0 \times 10^{18}$ yr ($0\nu 2\varepsilon$) | 40 |
| $^{114}\text{Cd}\rightarrow^{114}\text{Sn}$ | $\text{CdWO}_4$ | $\geq 1.3 \times 10^{18}$ yr ($2\nu 2\beta^-$) | 40 |
|                     | $\text{CdWO}_4$ | $\geq 1.1 \times 10^{21}$ yr ($0\nu 2\beta^-$) | 40 |
| $^{116}\text{Cd}\rightarrow^{116}\text{Sn}$ | $^{116}\text{CdWO}_4$ | $= 2.69 \pm 0.02(\text{stat.}) \pm 0.14(\text{syst.}) \times 10^{19}$ yr ($2\nu 2\beta^-$) | 41 |
|                     | $^{116}\text{CdWO}_4$ | $\geq 2.4 \times 10^{23}$ yr ($0\nu 2\beta^-$) | 41 |
| $^{136}\text{Xe}\rightarrow^{136}\text{Ba}$ | $\text{Xenon-loaded liquid scintillator}$ | $= 2.21 \pm 0.02(\text{stat.}) \pm 0.07(\text{syst.}) \times 10^{24}$ yr ($2\nu 2\beta^-$) | 25 |
| $^{130}\text{Ba}\rightarrow^{130}\text{Xe}$ | $\text{BaF}_2$ | $\geq 1.4 \times 10^{17}$ yr ($0\nu \beta^+$) | 42 |
| $^{136}\text{Ce}\rightarrow^{136}\text{Ba}$ | $\text{CeCl}_3$ | $\geq 3.2 \times 10^{16}$ yr ($2\nu 2\kappa$) | 43 |
| $^{160}\text{Gd}\rightarrow^{160}\text{Dy}$ | $\text{Gd}_2\text{SiO}_5(\text{Ce})$ | $\geq 1.9 \times 10^{19}$ yr ($2\nu 2\beta^-$) | 44 |
|                     | $\text{Gd}_2\text{SiO}_5(\text{Ce})$ | $\geq 1.3 \times 10^{21}$ yr ($0\nu 2\beta^-$) | 44 |
| $^{181}\text{W}\rightarrow^{181}\text{Hf}$ | $\text{CaWO}_4$ | $\geq 3.1 \times 10^{19}$ yr ($2\nu 2\kappa$) | 29 |
|                     | $\text{CaWO}_4$ | $\geq 9.4 \times 10^{18}$ yr ($0\nu 2\varepsilon$) | 29 |
| $^{186}\text{W}\rightarrow^{186}\text{Os}$ | $\text{ZnWO}_4$ | $\geq 2.3 \times 10^{19}$ yr ($2\nu 2\beta^-$) | 32 |
|                     | $^{116}\text{CdWO}_4$ | $\geq 1.1 \times 10^{21}$ yr ($0\nu 2\beta^-$) | 45 |
Large-scale scintillator-based projects to search for neutrinoless double $\beta$ decay with sensitivity on the level of the inverted hierarchy of the neutrino masses have been proposed [46, 47, 48]. In this regard, it is of note that the SuperNEMO double $\beta$ decay project [49] intents to utilize a large amount of plastic scintillators for the calorimeter of the detector [50].

Cryogenic scintillation bolometers (see e.g. [51, 52, 53, 54, 55]), with a typical energy resolution of a few keV and potentially with strong reduced background, look perspective technique for future $0\nu2\beta$ decay experiments able to explore the full range of the inverted hierarchy of the neutrino mass (half-life sensitivity on the level of $10^{26} - 10^{28}$ years) [56, 57, 58]. At present ZnSe [59, 60, 61, 33], Li$_2$MoO$_4$ [62, 35] and CaMoO$_4$ [63, 64], CdWO$_4$ [65, 66, 67] are considered as the most promising materials for high sensitivity scintillating-bolometers $2\beta$ decay experiments. The large-scale experiments intend to use $10^2 - 10^3$ kg of highly radiopure scintillators enriched in the isotopes of interest. It should be mentioned R&D of other crystal scintillators containing molybdenum: ZnMoO$_4$ [68, 69, 35], Li$_2$Zn$_2$(MoO$_4$)$_3$ [70], Li$_2$Mg$_2$(MoO$_4$)$_3$ [71], Na$_2$MoO$_2$O$_7$ [72], Sr$_2$MoO$_4$ [73].

The double beta decay projects require as much as possible low, in ideal case zero, background of a detector in a region of interest. The most dangerous radionuclides for $2\beta$ experiments are $^{226}$Ra and $^{228}$Th, since their daughters ($^{214}$Bi and $^{208}$Tl) have large energies of decay: $Q_\beta = 3270$ keV and $Q_\beta = 4999$ keV, respectively. Potassium typically contributes to the energies below 1461 keV. However, $^{40}$K can produce background hampering $2\nu2\beta$ measurements. Presence of cosmogenic radioactivity should be also controlled and decreased as much as possible. A reachable (and measurable with present instrumentation) level of a few $\mu$Bq/kg in crystal scintillators is discussed now (see, e.g. [68, 35, 74, 75, 76]).

2.2 Dark matter

There is an evidence for a large amount of invisible (dark) matter in the Universe which reveals itself only through gravitational interaction. Weakly interacting massive particles (WIMPs), in particular neutralino, predicted by the Minimal Supersymmetric extensions of the Standard Model, are one of the many possible candidates of dark matter [77, 78, 79, 80, 81]. In direct detection investigation the annual modulation signature is a powerful tool because it is independent on the nature of the dark matter candidate. It is exploited e.g. by DAMA [82] with highly radiopure NaI(Tl) crystal scintillators.

In the case of the WIMP scenarios, WIMPs can be detected due to their scattering on nuclei producing low energy nuclear recoils. An extremely low counting rate (less than several counts kg$^{-1}$ d$^{-1}$) and small energy of recoil nuclei (below 100 keV) are expected in experiments to search for the WIMPs. Direct methods of WIMP detection are based on registration of ionization, scintillation or heat release caused by recoil nucleus embedded in the material of the detector; the nucleus could be in an excited state. At present, most sensitive direct experiments apply a variety of detection techniques for WIMP search: semiconductor detectors [83, 84, 85, 86, 87], noble gases based detectors [88, 89, 90, 91, 92, 93, 94, 95], bubble chambers [96], cryogenic bolometers [97, 98, 99]. Crystal scintillators are applied in conventional scintillation detectors [100, 101, 102, 82, 103] and in cryogenic scintillating bolometers, that use simultaneous registration of heat and light signals from crystal scintillators to reject background caused by electrons [104, 105]. There are several dark matter experiments in preparation using sodium

\footnote{The decay energy of the most promising $2\beta$ nuclei is $\sim 2 - 3$ MeV.}
iodine crystal scintillators as conventional room temperature scintillation detectors [106, 107, 108, 109, 110] and low-temperature scintillating bolometers [111]. Utilization of undoped CsI crystal scintillator as low-temperature scintillating bolometer is considered in Ref. [112]. High scintillation efficiency CaI$_2$ crystal scintillators (absolute light output $\sim 106,000$ photon/MeV) were recently proposed as WIMP detectors aiming at decreasing the energy threshold [113, 114]. Anisotropic ZnWO$_4$ crystal scintillator is proposed to search for directionality of dark matter signals [115, 116].

Radioactive contamination of target scintillation crystals plays a key role to decrease background in the experiments. Counting rate of a few counts kg$^{-1}$ d$^{-1}$ in the energy interval up to $\sim 20$ keV is typical in the present scintillator-based dark matter experiments [102, 82, 105]. The radioactive contamination of crystal scintillators used in dark matter experiments by potassium, uranium, radium, thorium and their daughters limits the experiments sensitivity. Besides, primordial ($^{87}$Rb, $^{113}$Cd, $^{115}$In, $^{138}$La, $^{176}$Lu, $^{187}$Re), cosmogenic (we refer reader to the review [117]) and artificial ($^{60}$Co, $^{134}$Cs, $^{137}$Cs, etc.) $\beta$ active nuclides can produce background in scintillation dark matter experiments too. It should be stressed that presence of these radionuclides in crystals on the levels significant for dark matter experiments can be detected in practice only under extremely low background conditions with high detection efficiency (actually, in the course of dark matter experiments).

2.3 Measurements of neutrino fluxes

Measurements of solar and reactor neutrino fluxes allow to refine our understanding of neutrino properties, in particular, to determine parameters of the Pontecorvo-Maki-Nakagawa-Sakata matrix and to test the Mikheyev-Smirnov-Wolfenstein effect [5, 118, 119, 120]. These experiments require large-volume detectors (tens - thousands tons) with an extremely low level of radioactive contamination ($\sim$nBq/kg). Especially purified liquid scintillators are used for the real-time measurements of the solar neutrino flux in the Borexino experiment [121] and to measure anti-neutrino flux from distant nuclear reactors in the KamLAND detector [122].

Low radioactive liquid scintillation detectors are used in experiments to measure antineutrino fluxes from nuclear reactors [123]. There are several projects of large scale experiments with reactor neutrinos. The Daya Bay [124], Double Chooz [125], and RENO [126] experiments utilized gadolinium-doped liquid scintillator to measure the value of the neutrino mixing angle $\theta_{13}$. The ambitious JUNO project intents to utilize 20 kton liquid scintillation underground detector aiming at determination of the neutrino mass hierarchy [127, 128]. It is expected that the intrinsic radiopurity of the scintillator should be better [129] to those reached in the Borexino and KamLAND detectors.

2.4 Search for hypothetical processes and particles

Scintillation detectors were used in a number of experiments to search for hypothetical processes beyond the Standard Model: decay of electron with violation of electric charge conservation, decay of nucleons and pairs of nucleons, charge non-conserving (CNC) beta decay, violation of the Pauli exclusion principle (PEP), search for magnetic momentum of neutrino, etc. Searches for the PEP violation were realized in [130, 131, 132] utilizing data of low background experiments with NaI(Tl) crystal scintillators. Data of the DAMA experiment have also been used to search for CNC transitions in $^{23}$Na and $^{127}$I nuclei [133], searches for nucleons decay to invisible
channels [134], instability of electron [131] [135], solar axions [136]. Large mass and ultra-low level of radioactive background of the Borexino detector have allowed to establish new limits on processes of decay of nucleons into invisible channels (for example, with emission of only neutrinos) [137], on magnetic momentum of neutrino [138], to search for solar axions [139, 140] and for the PEP violation [141]. Invisible nucleons decays were searched for also in the SNO and KamLAND experiments [142, 143]. In work [144] search for activity with electric charge non-conservation was realized in the experiment with LaCl3(Ce) crystal scintillator.

2.5 Investigation of rare α and β decays

Crystal scintillators, both as ordinary scintillation detectors and as cryogenic scintillating bolometers, are successfully used to investigate rare α and β decays. The half-life and the spectrum shape of the fourth-forbidden β decay of 113Cd were measured with the help of CdWO4 crystal scintillators [145, 146, 147]. In the experiment [147] the 113Cd half-life was determined with the highest to-date accuracy \( T_{1/2} = (8.04 \pm 0.05) \times 10^{15} \text{ yr} \) [148] and 115In \( T_{1/2} = 4.41(26) \times 10^{14} \text{ yr} \) [149]. Recently YVO4 scintillating bolometer was identified as a promising tool for investigation of rare \( (T_{1/2} \approx 10^{17} \text{ yr}) \) β− and EC decays of 50V [150]. In 1960 Beard and Kelly have utilized small CaWO4 and CdWO4 crystals in low background experiments to search for alpha activity of natural tungsten [151]. However, even more sensitive experiment with CdWO4 crystal scintillator did not allow to observe α decays of tungsten [152]. The first indication on the decay of 180W with a half-life \( T_{1/2} = 1.1 \times 10^{18} \text{ yr} \) was obtained with the help of enriched 116CdWO4 crystal scintillator [153]. This observation was confirmed with CaWO4 crystals (as cryogenic scintillating bolometer [154] and room temperature scintillation detector [155]) and ZnWO4 crystal scintillator [156]. The alpha activity of bismuth (209Bi, considered before as the heaviest stable element in the nature) with the half-life \( T_{1/2} = (1.9 \pm 0.2) \times 10^{19} \text{ yr} \) has been detected with the help of BGO cryogenic scintillator [157]. In the same approach, decay of 209Bi to the first excited level of 205Tl was also observed [158]. An indication on rare α activity of natural europium (151Eu) obtained in the low background experiment by using CaF2(Eu) crystal scintillator [159] was confirmed with Li6Eu(BO3)3 crystal scintillator operated as a scintillating bolometer [160]. Half-life limits on α decays of Pb isotopes were obtained with PbWO4 scintillating bolometer grown from ancient Roman lead with low 210Pb activity [161]. ZnWO4 scintillating bolometer doped with enriched 148Sm was used for more precise measurement of 148Sm half-life as \( T_{1/2} = 6.4^{+1.2}_{-1.3} \times 10^{15} \text{ yr} \) [162]. An interesting study of the L/K electron capture ratio in the decay of 207Bi decay to the 1633 keV excited level of 207Pb was realized with a BGO scintillating bolometer contaminated by 207Bi [163]. Rare (probability of \( \sim 10^{-9} \)) emission of e+e− pairs in α decay of 241Am was measured with NaI(Tl) scintillators in Ref. [164].

3 Experimental methods to measure radioactive contamination of scintillators

Methods of determination of radioactive contaminants in scintillators could be classified as direct, when characteristic radioactivity of specific isotope is detected, and indirect, which give quantitative conclusion on the presence of specific isotope on the basis of measurements of
contamination by the corresponding chemical element (by mass-spectrometry or fluorescence methods) or its daughters (by neutron activation analysis). The main characteristics of elements containing primordial radioactive isotopes are presented in Table 2. While indirect methods can be applied to any material, scintillators can measure their own internal radioactive contamination by themselves. This provides in general higher sensitivity, and further we will give more details on methods of analysis of radioactive contaminants in the data collected with scintillators.
Table 2: Elements containing primordial radioactive isotopes (except radium that is originated from $^{238}$U decay). Data on the half-lives of $\alpha$ and $\beta$ active isotopes are taken from [165] if other source is not referred; year is accepted as 1 yr = 365.2422 d; the isotopic abundances are from [166] (the abundance of $^{226}$Ra is assumed to be 100%); the atomic weights are from [167].

| Element | Radioactive isotope, decay modes | Isotopic abundance | Half-life (yr) | Activity in 1 g of element (Bq) | Mass of element (g) corresponding to 1 mBq activity of the radioactive isotope |
|---------|---------------------------------|-------------------|---------------|---------------------------------|---------------------------------|
| Potassium | $^{40}$K, $\alpha$, $\beta^-$ | 0.000117(1) | $1.248(3) \times 10^{10}$ | 31.7(3) | 3.15(3) $\times 10^{-5}$ |
| Calcium | $^{46}$Ca, $\beta^-$ | 0.00187(21) | $6.41(1) \times 10^{10}$ | 102 | 10.3(1) $\times 10^{6}$ |
| Vanadium | $^{51}$V, $\alpha$ | 0.00250(10) | $2.39(25) \times 10^{17}$ | 2.8(3) $\times 10^{6}$ | 352(40) |
| Germanium | $^{64}$Ge, $\beta^-$ | 0.0775(12) | $1.926(94) \times 10^{21}$ | 7.1(4) $\times 10^{6}$ | 1.41(7) $\times 10^{8}$ |
| Selenium | $^{82}$Se, $\beta^-$ | 0.0882(15) | $9.6(10) \times 10^{18}$ | 1.5(2) $\times 10^{-7}$ | 6.5(7) $\times 10^{4}$ |
| Rubidium | $^{87}$Rb, $\alpha$ | 0.2783(2) | $4.97(3) \times 10^{19}$ | 867(5) | 1.15(4) $\times 10^{-6}$ |
| Zirconium | $^{89}$Zr, $\beta^-$ | 0.0280(2) | $2.35(21) \times 10^{19}$ | 1.73(15) $\times 10^{-7}$ | 5.8(5) $\times 10^{4}$ |
| Molybdenum | $^{100}$Mo, $\beta^-$ | 0.0974(65) | $6.9(4) \times 10^{18}$ | 1.95(11) $\times 10^{-6}$ | 514.31 |
| Cadmium | $^{111}$Cd, $\beta^-$ | 0.12227(7) | $8.04(5) \times 10^{15}$ | 1.789(11) $\times 10^{-8}$ | 0.559(4) |
| | $^{116}$Cd, $\beta^-$ | 0.07512(54) | $2.69(14) \times 10^{19}$ | 3.28(17) $\times 10^{-7}$ | 3.05(16) $\times 10^{4}$ |
| Indium | $^{115}$In, $\beta^-$ | 0.95719(52) | $4.41(25) \times 10^{17}$ | 0.250(14) | 4.00(23) $\times 10^{-3}$ |
| Tellurium | $^{128}$Te, $\beta^-$ | 0.3174(8) | $2.0(3) \times 10^{24}$ | 1.65(25) $\times 10^{-11}$ | 6.1(9) $\times 10^{-7}$ |
| | $^{130}$Te, $\beta^-$ | 0.3408(62) | $8.2(6) \times 10^{20}$ | 4.3(3) $\times 10^{-8}$ | 2.32(19) $\times 10^{4}$ |
| Lanthanum | $^{138}$La, $\beta^-$ | 0.008881(71) | $1.03(1) \times 10^{14}$ | 0.82(11) | 1.218(15) $\times 10^{-4}$ |
| Neodymium | $^{144}$Nd, $\alpha$ | 0.23078(19) | $2.29(16) \times 10^{15}$ | 9.5(7) $\times 10^{-3}$ | 0.105(7) |
| | $^{150}$Nd, $\beta^-$ | 0.05638(28) | $9.34(0.66) \times 10^{18}$ | 5.5(4) $\times 10^{-7}$ | 1.81(3) $\times 10^{3}$ |
| Samarium | $^{147}$Sm, $\alpha$ | 0.1500(14) | $1.60(11) \times 10^{11}$ | 124.5(18) | 8.03(11) $\times 10^{-6}$ |
| | $^{148}$Sm, $\alpha$ | 0.1125(9) | $6.4(1) \times 10^{15}$ | 1.5(3) $\times 10^{-3}$ | 0.65(13) |
| Europium | $^{151}$Eu, $\alpha$ | 0.4718(6) | $4.6(1.2) \times 10^{15}$ | 9.0(24) $\times 10^{-6}$ | 111(29) |
| Gadolinium | $^{152}$Gd, $\alpha$ | 0.00203(8) | $1.08(8) \times 10^{14}$ | 1.56(27) $\times 10^{-8}$ | 0.64(10) |
| Lutetium | $^{171}$Lu, $\beta^-$ | 0.02599(13) | $3.64(35) \times 10^{10}$ | 54.0(6) | 1.853(20) $\times 10^{-5}$ |
| Hafnium | $^{174}$Hf, $\alpha$ | 0.0016(12) | $2.0(4) \times 10^{15}$ | 6.5(5) $\times 10^{-5}$ | 17(13) |
| Tungsten | $^{180}$W, $\alpha$ | 0.0012(1) | $1.8(2) \times 10^{18}$ | 4.8(5) $\times 10^{-8}$ | 2.08(23) $\times 10^{4}$ |
| Rhenium | $^{187}$Re, $\beta^-$ | 0.6260(5) | $4.53(7) \times 10^{11}$ | 1027(17) | 9.74(16) $\times 10^{-7}$ |
| Osmium | $^{188}$Os, $\alpha$ | 0.01596(4) | $2.0(11) \times 10^{15}$ | 6.4(2) $\times 10^{-4}$ | 1.8(12) |
| Platinum | $^{192}$Pt, $\alpha$ | 0.0001(2) | $4.97(16) \times 10^{11}$ | 0.0164(29) | 0.061(10) |
| Bismuth | $^{209}$Bi, $\alpha$ | 1 | $2.0(1) \times 10^{19}$ | 3.15(13) $\times 10^{-6}$ | 318(13) |
| Radium | $^{226}$Ra, $\alpha$ | 1 | $1600(7)$ | $3.658(16) \times 10^{10}$ | 2.734(12) $\times 10^{-14}$ |
| Thorium | $^{230}$Th, $\alpha$ | 0.0002(2) | $7.54(3) \times 10^{9}$ | 1.5(15) $\times 10^{6}$ | 6.6(66) $\times 10^{-9}$ |
| | $^{232}$Th, $\alpha$ | 0.9998(2) | $1.40(1) \times 10^{10}$ | 4071(29) | 2.456(18) $\times 10^{-7}$ |
| | $^{232}$U, $\alpha$ | 0.000054(5) | $2.433(6) \times 10^{9}$ | 1.22(12) $\times 10^{4}$ | 8.2(7) $\times 10^{-8}$ |
| | $^{238}$U, $\alpha$ | 0.00720(6) | $7.04(1) \times 10^{9}$ | 568.7(9) | 1.7585(29) $\times 10^{-6}$ |
| | $^{238}$U, $\alpha$ | 0.007204(10) | $4.468(6) \times 10^{9}$ | 12347(17) | 8.099(11) $\times 10^{-8}$ |
3.1 Indirect methods

Long living radioactive isotopes can be measured with the help of Inductively Coupled Plasma Mass Spectrometry (ICP-MS). Sensitivity of this method depends on measured matrix and on previous use of an apparatus. For instance, sensitivity of the spectrometers installed in the Gran Sasso underground laboratory of I.N.F.N. (Italy) to the solids and reagents involved in the TeO$_2$ crystals production process was on the level of $\sim (2 \times 10^{-10} - 2 \times 10^{-12})$ g/g for $^{232}$Th and $^{238}$U [178], which corresponds to activity of $^{232}$Th and $^{238}$U: $\sim (0.8 - 0.008)$ mBq/kg and $\sim (2 - 0.02)$ mBq/kg, respectively. Sensitivity of ICP-MS method to potassium is hampered due to interference of $^{39}$K with polyatomic species produced from argon used in the plasma source of ICP-MS devices [178]. The measured $^{39}$K concentrations in NaI powder for radiopure NaI(Tl) crystal scintillators production were $\sim (1 \times 10^{-8} - 2 \times 10^{-7})$ g/g, while the detection limit was on the level of $< 3 \times 10^{-9}$ g/g [178], that corresponds to $^{40}$K activity $\sim (0.3 - 6)$ mBq/kg and a detection limit $< 0.09$ mBq/kg. A higher sensitivity (a detection limit 0.016 mBq/kg of $^{40}$K in NaI matrix) was reported in [179]. The improved sensitivity was achieved thanks to the use of improved instrumentation, cool plasma operating conditions, and meticulously clean sample preparations. Accelerator mass-spectrometry allows to reach much higher sensitivity up to $\sim 10^{-17}$ g/g (potassium in liquid scintillator [180]). Unfortunately, mass-spectrometry is practically useless to measure contamination by $^{226}$Ra (daughter of $^{238}$U) and $^{228}$Th ($^{232}$Th) due to the rather short half-lives of these isotopes. At the same time, activities of these radionuclides (those daughters, $^{214}$Bi and $^{208}$Tl, are the most unfavorable background sources for double $\beta$ decay experiments) can be substantially different from $^{238}$U and $^{232}$Th due to broken secular equilibrium. The same can be said about $^{210}$Pb, daughter of $^{238}$U, one of the most troublesome contaminant radionuclides for dark matter experiments.

A very high sensitivity to $^{40}$K, $^{232}$Th and $^{238}$U on the level of nBq/kg was reached by using neutron-activation technique to measure contamination of liquid scintillator for the KamLAND experiment [181]. While neutron-activation analysis is very powerful tool to measure radioactive contamination in organic scintillators, the method is much less effective to examine inorganic materials due to possible activation of scintillation crystals matrix [182]. Comparison of the most sensitive indirect methods (mass spectrometry and neutron activation technique) with direct counting is discussed in [183].

It should be stressed that the various analytical methods used for analysis of chemical impurities in raw materials for crystal growth, like for instance Atomic Absorption Spectroscopy, X-ray Fluorescence, allow to estimate presence of long-living naturally occurring primordial radioactive elements too. It is worth to mention also Electron Microscope measurements to estimate presence of platinum in $^{116}$CdWO$_4$ crystal scintillators [153]. However, the sensitivity of these methods are much below of the ICP-MS sensitivity, not to say for direct counting methods [182, 184].

3.2 Direct detection

3.2.1 Low background measurements

Ultra-low background HP Ge $\gamma$ detectors can be used to measure radioactive contamination of scintillation crystals and materials for their production (see, for instance [185, 186, 191, 192, 193, 194, 195, 196, 197, 198, 187, 188, 189, 190]). This method provides typical sensitivity at the level of mBq/kg for $^{40}$K, $^{137}$Cs, $^{228}$Th, $^{226}$Ra and $^{227}$Ac (daughter of $^{235}$U), and somewhat
lower sensitivity to other U/Th daughters. However, this approach is useless to detect internal contamination by $\alpha$ and $\beta$ active isotopes if decay goes to the ground state of a daughter nucleus.\(^2\)

The highest sensitivity to measure internal contamination of scintillators can be achieved in low background measurements where the scintillator is operating as a detector. Such an approach provides high detection efficiency, especially for $\alpha$ and $\beta$ particles. A typical low background scintillation set-up (see, for instance, \[199, 200, 201, 202, 203, 144, 204, 205, 206, 207, 208, 209, 41\]) consists of scintillator, light-guide to shield the scintillator from radioactivity of photomultiplier tubes (typically, the most contaminated details of a low background scintillation set-up), passive shield. Background of the detector can be further suppressed by using of active shield counters surrounding the main detector, and anti-muon veto counters \[45, 195\]. Light-guides made of a scintillation material with different (relative to the main scintillation detector) scintillation decay time can serve as active anticoincidence detectors \[45, 195, 38\]. Continuous flushing of internal volume of a set-up by a radon-free gas (typically by nitrogen) allows to reduce background caused by radon \[205, 209\]. It is worth mentioning a possibility to reduce background further by using data on time of arrival and pulse-shape of scintillation signals (the methods will be considered in Sections \[3.2.3 and 3.2.4\]).

Below we will discuss response of a scintillation detector to $\gamma$ quanta, $\beta$ and $\alpha$ particles, pulse-shape discrimination, time-amplitude analysis, Monte-Carlo simulation of background components. These methods allow to describe background energy spectra accumulated with a scintillation counter and estimate radioactive contamination of the scintillator.

### 3.2.2 Response of scintillation detector to $\gamma$ quanta, $\beta$ and $\alpha$ particles

Knowledge of a detector response to $\gamma$ quanta (response function, dependence of energy resolution on energy) and $\alpha$ particles (energy dependence of $\alpha/\beta$ ratio\(^3\) and energy resolution) is necessary to interpret background of the detector.

Response function and dependence of energy resolution on energy of $\gamma$ quanta can be measured with $\gamma$ sources in a wide energy interval from a few keV (5.9 keV Mn K X-rays from $^{55}$Fe) up to 2615 keV ($\gamma$ quanta of $^{208}$Tl). Calibration with $\alpha$ sources is much more complicated task because energies of commonly used $\alpha$ sources lie in the energy region from $\sim 5.3$ to $\sim 8.8$ MeV ($^{228}$Th, $^{241}$Am, $^{244}$Cm, $^{252}$Cf). To calibrate detector at lower energies, $\alpha$ sources with absorbers can be used (see, for instance \[153, 205, 155, 210, 159\]). Response of scintillation detectors to $\alpha$ particles is non-linear. An example of $\alpha/\beta$ ratio dependence on energy is shown in Fig. 1, where the $\alpha/\beta$ ratio measured with a CaWO$_4$ crystal scintillator is presented. As the quenching of the scintillation light caused by $\alpha$ particles is due to the higher ionization density, such a behavior of the $\alpha/\beta$ ratio can be explained by the energy dependence of ionization density of particles \[211, 212\]. See recent review \[213\] on $\alpha/\beta$ ratio in different scintillators.

Alpha peaks from internal contamination of scintillators allow to extend the interval of $\alpha$ particles energies (see Fig. 1 and Fig. 2). In addition, $\alpha$ peaks from internal contamination provide an important test of calibration measurements with external sources. It should be

\(^2\)Beta activity can be detected by HP Ge detectors via registration of bremsstrahlung, however the sensitivity in this case is much lower due to low efficiency and absence of a clear signature (like peaks in $\gamma$ spectra).

\(^3\)Here we define the “$\alpha/\beta$ ratio” as the ratio of $\alpha$ peak position measured in the $\gamma$ energy scale to the energy of $\alpha$ particles. As usual, a detector energy scale is measured with $\gamma$ sources, thus the notation “$\alpha/\beta$ ratio” is more adequate. However, because $\gamma$ rays interact with matter by means of the energy transfer to electrons, in the present paper we are using a more traditional term “$\alpha/\beta$ ratio”.
Energy of α particles (MeV)

0 2 4 6 8

Figure 1: The energy dependence of the α/β ratio measured with CaWO₄ crystal scintillator [155]. The points obtained by irradiation of the scintillator by external α particles are shown by circles, while the points obtained by analysis of α peaks from the internal contamination of the crystal by α active nuclides are drawn by triangles.

stressed that analysis of internal α peaks is only practical method to measure response to α particles for encapsulated scintillation detectors produced from highly hygroscopic materials, like NaI(Tl), LaCl₃(Ce), LaBr₃, etc.

In some scintillation crystals with anisotropic structure, α/β ratio depends on the direction of α particles relatively to crystal axes. Such an effect was observed in anthracene [214], stilbene [215], and CdWO₄ [153], ZnWO₄ [210], and MgWO₄ [216] crystal scintillators (see Fig. 2 where dependence of α/β ratio on direction of α irradiation relatively to crystal axes of CdWO₄ scintillator is presented). It leads to some worsening of energy resolution of these detectors to α particles [153, 32].

Scintillation signals for γ quanta can be quenched in comparison to β particles. E.g. there is an indication of scintillation light-efficiency quenching in CdWO₄ scintillator for γ quanta in comparison to electrons that resulted in a higher $Q(γ)$ value of $^{113}$Cd [146, 147]: $Q(γ) = 337$ keV and $Q(β) = 345$ keV, respectively. These values are substantially larger than the Table value $Q(β) = 323.83(27)$ keV [217]. The quenching can be explained by the non-proportionality in the scintillation response of CdWO₄ crystal scintillators significant for energies below $\sim 0.1$ MeV [218]. Because of distribution of γ quanta energy between two and more electrons (due to the multiple Compton scattering), γ peaks should be shifted to lower energies in “electron” energy scale.$^4$ Organic scintillators, particularly liquid scintillators, have significant non-proportionality of scintillation response. The effect was observed in liquid scintillators used for neutrino experiments Borexino [221] and Double Chooz [222]. The effect of γ peaks shift in liquid scintillators is significant: e.g. the position of the 1461 keV γ peak of $^{40}$K in the scintillator used in the Borexino detector is shifted to 1360 keV relatively to the energy deposited for electrons [137].

$^4$This effect also leads to worse energy resolution of scintillation detectors for γ quanta [219, 220].
Figure 2: Energy dependence of the $\alpha/\beta$ ratio on energy measured with $^{116}$CdWO$_4$ crystal scintillator. The $\alpha/\beta$ ratio depends on direction of irradiation relatively to crystal axes denoted as dir. 1 (along to the crystal axis [010]), dir. 2 ([001]) and dir. 3 ([100]) [153]. The points obtained by irradiation of the scintillator by external $\alpha$ particles in the directions parallel to the axes of the crystal are shown by circles, while the points obtained by analysis of $\alpha$ peaks from internal contamination of the scintillator by $\alpha$ active nuclides are drawn by triangles.

### 3.2.3 Time-amplitude analysis

Data on the energy and arrival time of events can be used to select fast decay chains from the $^{232}$Th, $^{235}$U and $^{238}$U families. The method of time-amplitude analysis is described in detail in [223, 224, 44, 45, 208]. For instance, the following sequence of $\alpha$ decays from the $^{232}$Th family can be selected:

\[
^{224}\text{Ra} \left( Q_\alpha = 5789 \text{ keV}; \ T_{1/2} = 3.632 \text{ d} \right) \rightarrow \ ^{220}\text{Rn} \left( Q_\alpha = 6405 \text{ keV}; \ T_{1/2} = 55.6 \text{ s} \right) \rightarrow \ ^{216}\text{Po} \\
\left( Q_\alpha = 6906 \text{ keV}; \ T_{1/2} = 0.145 \text{ s} \right) \rightarrow \ ^{212}\text{Pb}.
\]

These radionuclides are in equilibrium with $^{228}$Th ($^{232}$Th family). As an example, the results of the time-amplitude analysis of data accumulated in the low background experiment to search for $2\beta$ decay of $^{116}$Cd with the help of $^{116}$CdWO$_4$ crystal scintillators are shown in Fig. 3. The obtained $\alpha$ peaks (the $\alpha$ nature of events was confirmed by the pulse-shape analysis described in Section 3.2.4) as well as the distributions of the time intervals between events, are in a good agreement with those expected for the $\alpha$ decays of the $^{224}$Ra $\rightarrow$ $^{220}$Rn $\rightarrow$ $^{216}$Po $\rightarrow$ $^{212}$Pb chain.

Similarly the following fast sequence of $\beta$ and $\alpha$ decays:

\[
^{214}\text{Bi} \left( Q_\beta = 3269 \text{ keV}; \ T_{1/2} = 19.9 \text{ m} \right) \rightarrow \ ^{214}\text{Po} \left( Q_\alpha = 7834 \text{ keV}; \ T_{1/2} = 164.3 \mu\text{s} \right) \rightarrow \ ^{210}\text{Pb}
\]

(in equilibrium with $^{226}$Ra from the $^{238}$U family) can also be selected with the help of time-amplitude analysis. In Fig. 4 one can see the energy spectra and time distributions of the sequence selected from the data accumulated in the low background experiment to search for $2\beta$ decay of $^{160}$Gd with the help of gadolinium orthosilicate ($\text{Gd}_2\text{SiO}_5$:Ce, GSO) scintillator [44]. In addition, Fig. 4 illustrates a possibility to detect another short chain:
Figure 3: The $\alpha$ peaks of $^{224}$Ra, $^{220}$Rn, and $^{216}$Po selected by the time-amplitude analysis from the data accumulated during 14745 h with $^{116}$CdWO$_4$ detector [153]. (Insets) The time distributions between the first and second (and between second and third) events are presented together with exponential fits. Obtained half-lives of $^{220}$Rn and $^{216}$Po ($57^{+9}_{-7}$ s and $0.140(8)$ s, respectively) are in a good agreement with the table values ($55.6(1)$ s and $0.145(2)$ s, respectively [165]).
Figure 4: The energy spectra of the sequence of $\beta$ and $\alpha$ decays in the decay chain $^{214}$Bi $\rightarrow$ $^{214}$Po $\rightarrow$ $^{210}$Pb ($^{238}$U family) which were selected with the help of the time-amplitude analysis of 8609 h data accumulated with GSO scintillator [44]. The peak with the energy in $\gamma$ scale $\approx$ 1.8 MeV is related with the decays of $^{219}$Rn from the chain $^{219}$Rn $\rightarrow$ $^{215}$Po $\rightarrow$ $^{211}$Pb ($^{235}$U family). In the insert: the distribution of the time intervals between the first and second events together with its fit (solid line) by the sum of exponent with $T_{1/2} = 129^{+90}_{-40}$ $\mu$s (a table $^{214}$Po value is $T_{1/2} = 164.3$ $\mu$s; dashed line) and exponent with $T_{1/2} = 1.78$ ms corresponding to decays of $^{215}$Po from the chain $^{219}$Rn $\rightarrow$ $^{215}$Po $\rightarrow$ $^{211}$Pb (dotted line).

$^{219}$Rn ($Q_\alpha = 6946$ keV; $T_{1/2} = 3.96$ s) $\rightarrow$ $^{215}$Po ($Q_\alpha = 7526$ keV; $T_{1/2} = 1.781$ ms) $\rightarrow$ $^{211}$Pb.

Radionuclide $^{219}$Rn is in equilibrium with $^{227}$Ac from the $^{235}$U family. In this case the events of $^{214}$Po and $^{215}$Po decays are superimposed (see Fig. 4). Nevertheless activities of $^{226}$Ra and $^{227}$Ac can be calculated separately thanks to possibility to distinguish between the broad $\beta$ spectrum of $^{214}$Bi and $\alpha$ peak of $^{219}$Rn.

### 3.2.4 Pulse-shape discrimination

Most of scintillators have slightly different decay kinetic for $\beta$ particles ($\gamma$ quanta) and $\alpha$ particles. It allows to discriminate these particles, and therefore, to estimate activity of $\alpha$ and $\beta$ active nuclides separately. Different methods can be used to realize pulse-shape discrimination. We would like to refer to the optimal filter method proposed by Gatti and De Martini [225], and developed in [226] for CdWO$_4$ crystal scintillators, and then successfully applied to many scintillators [205, 229, 210, 153, 155, 159, 227, 228, 216]. In the optimal filter method, a shape indicator ($SI$) - a numerical characteristic - can be calculated for each scintillation event produced by a scintillator:

$$SI = \frac{\sum [f(t_k) \times P(t_k)]}{\sum f(t_k)},$$

where the sum is over time channels $k$ from the origin of the pulse up to a certain time; $f(t_k)$
is the digitized amplitude of the signal (at the time \( t_k \)). The weight function \( P(t_k) \) is defined as:

\[
P(t_k) = \frac{f_\alpha(t_k) - f_\gamma(t_k)}{f_\alpha(t_k) + f_\gamma(t_k)},
\]

where \( f_\alpha(t_k) \) and \( f_\gamma(t_k) \) are the reference pulse shapes for \( \alpha \) particles and \( \gamma \) quanta. To realize the optimal filter method, the pulse shapes for \( \alpha \) particles and \( \gamma \) quanta should be studied.

The distributions of the shape indicators have Gaussian shape. Therefore, the figure of merit \( (FOM) \) – a measure of discrimination ability – can be calculated using the following expression proposed by Gatti and De Martini:

\[
FOM = \frac{|SI_\alpha - SI_\gamma|}{\sqrt{\sigma_\alpha^2 + \sigma_\gamma^2}}.
\]

One can see an illustration of pulse-shape discrimination by using the optimal filter method in Fig. 5 where the scatter plot of the shape indicator versus energy is shown for the data measured with a CaWO\(_4\) crystal scintillator [230]. Energy spectrum of \( \alpha \) events selected from the data measured with the CaWO\(_4\) crystal over 1734 h is presented in Fig. 6 while an energy spectrum of \( \beta(\gamma) \) events is shown in Fig. 7.

The mean time method is also widely used to discriminate \( \beta(\gamma) \) and \( \alpha \) particles in scintillation detectors (see e.g. [187, 202, 208, 228]). The following formula can be applied to calculate the mean time parameter \( \langle t \rangle \) for each pulse:
**Figure 6:** Energy spectrum of $\alpha$ events selected by the pulse-shape analysis from background data measured over 1734 h with CaWO$_4$ detector [230]. (Inset a) The same spectrum but scaled up. It is well reproduced by the model, which includes $\alpha$ decays of nuclides from $^{232}$Th and $^{238}$U families. (Inset b) Low energy part of the spectrum where an $\alpha$ peak of $^{147}$Sm is clearly visible.

**Figure 7:** Energy spectrum of $\beta(\gamma)$ events selected by the pulse-shape analysis technique from the background data measured over 1734 h with CaWO$_4$ detector [230]. The distribution is described by the $\beta$ spectra of $^{210}$Bi, $^{214}$Pb, $^{211}$Pb, $^{234m}$Pa, and $^{214}$Bi. (Inset) In the low energy region, the background (measured over 15.8 h) is caused mainly by decay of $^{210}$Pb.
Figure 8: The energy distributions for the fast sequence of β\(^{(212}\text{Bi, } Q_\beta = 2252 \text{ keV})\) and α\(^{(212}\text{Po, } E_\alpha = 8785 \text{ keV, } T_{1/2} = 0.299 \text{ s})\) decays selected by the pulse-shape analysis from the background data obtained in the experiment with cadmium tungstate crystal scintillators \[153\]. (Upper inset) The time distribution of intervals (\(\Delta t\)) between β and α signals. (Lower inset) Example of such an event in the CdWO\(_4\) scintillator.

\[
\langle t \rangle = \sum (f(t_k) \times t_k) / \sum f(t_k),
\]

where the sum is over time channels \(k\), starting from the origin of pulse and up to a certain time. The distributions of parameters \(\langle t \rangle\) for α and β(γ) signals are also well described by Gaussian functions. Thus, the same measure (FOM, see Equation 3) can be used to estimate efficiency of the method. The optimal filter method provides slightly better pulse-shape discrimination in comparison with the mean time technique (see e.g. \[228\]). However, the mean time method is easier to apply because it does not require the construction of a weight function, which requires knowledge of scintillation signals pulse-shape.

It should be stressed that the pulse-shape of scintillation signals for α particles depends on energy. In some scintillators with anisotropic properties pulse-shape also depends on direction of α irradiation relatively to the crystal axes. As in the case with the α/β ratio, such a dependence was observed in CdWO\(_4\) \[153\], ZnWO\(_4\) \[210\], and MgWO\(_4\) \[216\] crystal scintillators.

Pulse-shape analysis can also be applied to the very fast sequence of decays from the \(^{232}\text{Th}\) family:

\(^{212}\text{Bi (} Q_\beta = 2252 \text{ keV; } T_{1/2} = 60.55 \text{ m}) \rightarrow ^{212}\text{Po (} Q_\alpha = 8954 \text{ keV; } T_{1/2} = 0.299 \mu\text{s}) \rightarrow ^{208}\text{Pb.}\)

An example of such an analysis is presented in Fig. 8 where the β spectrum of \(^{212}\text{Bi, the α peak of } ^{212}\text{Po and the distribution of the time intervals between the first and the second pulses selected from the data of low background experiment \[153\] are depicted. This method is more effective with fast scintillators.\)
Figure 9: The background spectrum of the GSO detector for 0.969 yr kg of exposure (points) and the model of background (solid line) obtained by the fit of the data in the 60 – 3000 keV energy interval [44]. The most important internal ($^{40}$K, sum of $^{235}$U and $^{238}$U, $^{232}$Th) and external ($\gamma$ radiation from the photomultiplier tube) components of background are shown. A peak at the energy $\approx 420$ keV is due to the $\alpha$ activity of $^{152}$Gd, while the background in the energy interval 0.8 – 2 MeV is caused mainly by $\alpha$ decays of U/Th daughters.

### 3.2.5 Simulation of background energy spectra

To estimate possible contamination of a scintillator, especially by $\beta$ active nuclides, one can fit an energy spectrum by Monte Carlo simulated models. As an example, the fit of the low background energy spectrum accumulated with GSO crystal scintillator in the experiment to search for $2\beta$ decay of $^{160}$Gd [44] is presented in Fig. 9. The models of background were simulated with the help of the GEANT package [231, 232, 233]. For models presented in Fig. 9 an event generator DECAY0 [234] was used. This generator allows to take into account a number and types of emitted particles, their energies, directions of movement and times of emission. GEANT and EGS4 [235] packages are the most widely used to simulate background of scintillation detectors.

### 3.2.6 Low-temperature scintillating bolometers

An extremely high sensitivity to $\alpha$ radioactive contamination of crystal scintillators can be reached by operation of a scintillator as low temperature scintillating bolometer thanks to a very high energy resolution and particle discrimination capability [51, 52, 53, 55]. The discrimination can be realized by analysis of heat and scintillation signals from a crystal scintillator (by using a different scintillation yield for ions [212], in particular for $\alpha$ particles) or by only thermal pulse profile analysis [236].

The capability of low-temperature scintillating bolometers to screen $\alpha$ radioactive contamination of crystal scintillators is illustrated in Fig. 10, where $\alpha$ spectra gathered with two
Figure 10: (a) Energy spectrum of α events selected by pulse-shape discrimination from the data acquired by 589 g $^{116}$CdWO$_4$ scintillation detector at the Gran Sasso underground laboratory over 1727 h together with the model which includes α active nuclides of $^{238}$U and $^{232}$Th families [195]. (b) Energy spectrum of α events measured by 34.5 g $^{116}$CdWO$_4$ scintillating bolometer in a surface laboratory over 250 h [67]. An advantage of the high energy resolution and particle discrimination capability of the low-temperature scintillating bolometer is clearly visible.

samples of $^{116}$CdWO$_4$ scintillator operated as a conventional scintillation detector and as a cryogenic scintillating bolometer are presented. A small sample of $^{116}$CdWO$_4$ crystal (34.5 g) was measured over 250 h in a surface set-up [67]. The achieved sensitivity on the level of $\sim 0.1$ mBq/kg for $^{232}$Th, $^{235}$U and $^{238}$U (and their α active daughters) competes with results of the long time measurements (1727 h) with much larger mass $^{116}$CdWO$_4$ crystal scintillator (589 g) by using ultra-low background set-up deep underground at the Gran Sasso underground laboratory [195].

4 Data on radioactive contamination of scintillators

Data on radioactive contamination of different scintillators are presented in Tables 3-13. The main sources of radioactive contamination of scintillation materials are naturally occurring radionuclides of the $^{232}$Th and $^{238}$U families, and $^{40}$K. Activity of $^{235}$U with daughters is observed in some materials too. It should be stressed that the secular equilibrium of the U/Th chains is usually broken in scintillation materials. It means that the activities of $^{238}$U, $^{230}$Th, $^{226}$Ra, $^{210}$Pb in the $^{238}$U family should be considered separately. The activity of $^{210}$Po should be also reported separately if a scintillation material was produced recently in comparison to the half-
Figure 11: Time dependence of $^{210}$Po activity in CaWO$_4$ crystal scintillator [196] (circles). The data is approximated by a function, describing the increase in $^{210}$Po due to the decay of $^{210}$Pb assuming essentially lower amount of $^{210}$Po after the crystal growth (solid line). The $^{210}$Po activity is beginning to decline after $\sim 2.2$ yr due to decay of $^{210}$Pb ($T_{1/2} = 22.2$ yr).

life of $^{210}$Po ($T_{1/2} = 138.376(2)$ d). A time behaviour of $^{210}$Po activity in a CaWO$_4$ crystal scintillator, in the case of essentially lower amount of $^{210}$Po after the crystal growth, is shown in Fig. 11. Similarly, contaminations by $^{232}$Th, $^{228}$Ra and $^{228}$Th from the $^{232}$Th family, and activities of $^{235}$U, $^{231}$Pa and $^{227}$Ac from the $^{235}$U family can be different. As an example of a strong disequilibrium of the $^{232}$Th chain we can refer the CeF$_3$ crystal scintillator where the activity of $^{232}$Th exceeded the activity of $^{228}$Th by a factor 18 [205]. As for the $^{238}$U chain, its equilibrium is strongly broken e.g. in CaWO$_4$ [155] where the activities of $^{238}$U, $^{226}$Ra and $^{210}$Po relate as 1 : 0.4 : 21. Both the $^{232}$Th and $^{238}$U chains were found strongly broken in BaF$_2$ crystal scintillator, where radium accumulation ($^{226}$Ra from the $^{238}$U chain, and $^{228}$Ra from the $^{232}$Th chain) in the crystal was observed [244]. For this reason a reference date should be given for activities of relatively short living isotopes $^{210}$Po, $^{228}$Th, $^{228}$Ac.
Table 3: Radioactive contamination of organic scintillators.

| Chain | Source | Activity (mBq/kg) in sample, Reference |
|-------|--------|----------------------------------------|
|       |        | Liquid scintillators |                       | Plastic scintillators |                       |                       |
|       |        | Borexino, 278 t | KamLAND | Polystyrene-based scintillator | EJ-200 | NE-102A |
|       |        | Phase Ia | Phase II | [238] | [239] | [240] | [241] |
| $^{14}$C |  | 0.445(10) | 0.445(10) | 0.66(16) |                       |                       |                       |
| $^{35}$Ar |  | < 0.005 |                       | 1.5(5) | 2.2(7) |                       |                       |
| $^{40}$K |  | $< 1.1 \times 10^{-7}$ | $< 1.1 \times 10^{-7}$ | 3.4(3) \times 10^{-4} |                       |                       |
| $^{85}$Kr |  | $3.5(6) \times 10^{-6}$ | $< 6 \times 10^{-7}$ | 0.88(2) |                       |                       |
| $^{238}$Th | $^{232}$Th | 1.5(3) \times 10^{-8} | $< 4 \times 10^{-9}$ | 3.35(20) \times 10^{-7} |                       |                       |
| $^{226}$Ra |  | 0.4(1) | 9.2(4) |                       |                       |                       |
| $^{228}$Th |  | 0.5(1) | 9.3(3) |                       |                       |                       |
| $^{268}$Pb |  | $< 0.00013$ |                       | 0.0015(4) mBq/m² | b |                       |
| $^{238}$U | $^{226}$Ra | 6.6(4) \times 10^{-8} | $< 10^{-9}$ | 2.33(13) \times 10^{-8} |                       |                       |
| $^{210}$Pb |  | 0.09(5) | 0.96(13) |                       |                       |                       |
| $^{210}$Po |  | 0.0584(11) |                       |                       |                       |                       |
| $^{210}$Bi |  | 2.3 \times 10^{-6} | 2.3(6) \times 10^{-6} |                       |                       |                       |
| $^{209}$Po |  | 8 \times 10^{-6} | 8 \times 10^{-6} |                       |                       |                       |

a The period before purification of liquid scintillator in 2010 – 2011 is called Phase I, the period after the purification is denoted as Phase II.

b Surface contamination.
Table 4: Radioactive contamination of fluorite crystal scintillators.

| Chain | Source | Activity (mBq/kg) in sample, Reference |
|-------|--------|---------------------------------------|
|       | LiF(W) | CaF₂(Eu) | SrF₂ | CeF₃ | BaF₂ |
|       | 553 g  | 370 g  | 0.29 kg, 25 samples | 53 g  | 49 g  | 734 g | 1.714 kg |
| ⁴⁰K   | < 5.1  | < 7   | < 330 |
| ⁶⁰Sr  | < 4    |        |
| ¹³⁷Cs | < 0.2  | < 0.3  |
| ¹³⁷Ba | < 60   |        |
| ¹³⁷La | 0.34(4)| < 80   |
| ¹⁷⁶Lu |        | < 20   |
| ¹⁵²Eu | 10(2)  |        |
| ²³²Th | 0.05(1)| 55(30) | 400  | < 4  |
| ²²⁸Ra | < 0.6  | 890(270) |
| ²²⁸Ac | < 1.6  |        |
| ²²⁸Th | 0.13(2)| 1010(10)| 1.35(6) × 10³ |
| ²²⁵Ra |        | 21(1) |
| ²²⁸Rn | 0.036(4) - 40.72(12) |
| ²³⁸Pu | < 0.6  |        |
| ²³⁵U  | < 40   |        | < 0.6 |
| ²³⁵U  |        | < 0.6  |
| ²³⁵Pu | < 50   |        | < 0.7 |
| ²²⁶Ac | 0.011(7)| 0.012(20) - 2.05(31) | < 20  | < 70 |
| ²³⁸U  | 0.06(1)| < 70  | < 0.2 |
| ²³⁴U  |        | < 60  |
| ²³⁰Th |        | < 60  |
| ²²⁶Ra | 1.3(2) | 496(2) | < 60  | 1400 | 7.8(1) × 10⁴ |
| ²¹⁴Pb | 3.3(8) |        |
| ²¹⁴Bi | 3.3(8) | 0.052(23) - 75.3(4) |
| ²¹⁰Pb |        | 0.99(1) × 10⁴ |
| ²¹⁰Po | 0.9(2) | 72(6)  | < 280 |
| Total α activity | 8(2) | 3400 |
Table 5: Radioactive contamination of Y$_3$Al$_5$O$_{12}$:Nd [YAG(Nd)] and Li$_6$Eu(BO$_3$)$_3$ crystal scintillators.

| Chain | Activity (mBq/kg) in sample, Reference | YAG(Nd) | Li$_6$Eu(BO$_3$)$_3$ |
|-------|----------------------------------------|---------|---------------------|
|       |                                        | 7.16 g  | 2.72 g              |
| $^{40}$K |                           | [227]     | [191]             |
| $^{60}$Co |                         | < 1500   |                     |
| $^{137}$Cs |                       | < 26     |                     |
| $^{147}$Sm |                      | < 81     |                     |
| $^{207}$Bi |                       | < 9      |                     |
| $^{152}$Eu |                      | 949(48)  |                     |
| $^{154}$Eu |                      | 212(35)  |                     |
| $^{232}$Th | $^{232}$Th |                     | 3.5  |
| $^{228}$Ac |                       | < 200    |                     |
| $^{212}$Pb |                       | < 250    |                     |
| $^{208}$Tl |                       | < 130    |                     |
| $^{238}$U | $^{226}$Ra |                     | 2.9  |
| $^{214}$Po |                       | < 170    |                     |
| $^{214}$Bi |                       | < 70     |                     |
| $^{210}$Po |                       | 6.2      |                     |
| Total $\alpha$ activity | | < 10 | |
Table 6: Radioactive contamination of ZnSe and Zn$^{82}$Se crystal scintillators.

| Chain | Source          | Activity (mBq/kg) in sample, Reference |
|-------|-----------------|----------------------------------------|
|       | ZnSe 431 g  | Zn$^{82}$Se ≈ 440 g, 3 samples | Zn$^{82}$Se 174 – 481 g, 26 samples |
| $^{232}$Th | $^{232}$Th   | 0.0172(46)  | < 0.005 – 0.013(4) | < 0.0005 – 0.009(1) |
| $^{228}$Th | $^{228}$Th   | 0.0111(37)  | 0.022(4) – 0.032(7) | 0.0023(8) – 0.027(2) |
| $^{238}$U  | $^{238}$U    | 0.0246(55)  | < 0.01 – 0.020(5)  | < 0.001 – 0.013(2) |
| $^{234}$U  | $^{234}$U    | 0.0178(33)  | 0.001(2) – 0.015(4) |
| $^{234}$U+$^{226}$Ra | 0.0178(33) | 0.023(5) – 0.042(7) |
| $^{230}$Th | $^{230}$Th   | 0.0246(55)  | 0.017(4) – 0.019(5) | < 0.002 – 0.016(2) |
| $^{226}$Ra | $^{226}$Ra  | 0.0178(33)  | 0.004(1) – 0.018(2) |
| $^{218}$Po | $^{218}$Po   | 0.020(5) – 0.024(5) | 0.003(1) – 0.020(2) |
| $^{210}$Pb | $^{210}$Pb   | 0.100(12) – 0.250(17) |
| $^{210}$Po | < 0.1        | --          | --                      |
Table 7: Radioactive contamination of NaI(Tl) crystal scintillators.

| Chain | Source             | Activity (mBq/kg) in sample, Reference |
|-------|--------------------|----------------------------------------|
|       | 3.5 kg             | 10.7 kg, 246, 247                      |
|       | 12.5 kg, 214       | 239, 230                               |
|       | 9.7 kg ×25         | 9.7 kg ×25                             |
|       | Ø72 × 72 mm        | 251, 252                               |
|       | DAMA/LIBRA         | 253, 251                               |
|       | PICO-LON           | 255                                   |
| 224   | UK DM              | 254                                   |
| 238   | ANAIS              | DM-Ice17                               |
| 238   | DAMA/LIBRA         | DM-Ice17                               |
| 238   | PICO-LON           | DM-Ice17                               |
| 238   | KIMS-NaI           | DM-Ice17                               |
| 238   | DM-Ice17           | DM-Ice17                               |
| 238   | SABRE              | DM-Ice17                               |
| 238   | COSINE-100         | DM-Ice17                               |
| 238   |                    | DM-Ice17                               |
| 3H    | 0.2                | 254                                   |
| 22Na  | < 0.09             | DM-Ice17                               |
| 24Na  | < 0.00026          | DM-Ice17                               |
| 40K   | 1.1 – 21           | DM-Ice17                               |
| 85Kr  | < 0.01             | DM-Ice17                               |
| 87Rb  | < 0.3              | DM-Ice17                               |
| 129I  | 0.94               | DM-Ice17                               |
| 129I  | 0.95(6)            | DM-Ice17                               |
| 232Th | 0.0007 – 0.004     | DM-Ice17                               |
| 232Th | 0.0085(5)          | DM-Ice17                               |
| 232Th | 0.002 – 0.24       | DM-Ice17                               |
| 232Th | 0.001              | DM-Ice17                               |
| 226Ra | 0.16               | DM-Ice17                               |
| 226Ra | 0.014              | DM-Ice17                               |
| 210Po | 0.02               | DM-Ice17                               |
| 210Po | 0.013(8)           | DM-Ice17                               |
| 238U  | < 0.005            | DM-Ice17                               |
| 238U  | 0.0044(7)          | DM-Ice17                               |
| 238U  | < 0.0005 – 0.52    | DM-Ice17                               |
| 234U  | 2.7 – 10           | DM-Ice17                               |
| 234U  | 0.0158(16)         | DM-Ice17                               |
| 226Ra | 0.045              | DM-Ice17                               |
| 226Ra | 0.2                | DM-Ice17                               |
| 210Po | 0.7 – 3.15         | DM-Ice17                               |
| 210Po | 0.0217(11)         | DM-Ice17                               |
| 210Po | 0.105(17)          | DM-Ice17                               |
| 210Po | 0.03 – 0.96        | DM-Ice17                               |
| 210Po | < 0.0002 – < 0.015 | DM-Ice17                               |
| 210Po | 0.017              | DM-Ice17                               |
| 210Po | 0.0044(7)          | DM-Ice17                               |
| 210Po | 0.0158(16)         | DM-Ice17                               |
| 210Po | < 0.0005 – 0.52    | DM-Ice17                               |
| 210Po | 0.093 – 0.96       | DM-Ice17                               |

Total α activity: 0.58 – 3.15
Table 8: Radioactive contamination of CsI(Tl) and CsI(Na) crystal scintillators.

| Chain | Source | Activity (mBq/kg) in sample, Reference |
|-------|--------|----------------------------------------|
|       |        | CsI(Tl)                                 | CsI(Na)                           |
|       | 2 kg ×93 | 6.6 kg [208] 93 | 4.55 kg, 3 samples [187] 59 | ≈ 2 kg [260] |
|       | TEXONO | KIMS | China Dark matter Experiment | |
| 40K   |        | 17(16) | |
| 87Rb  |        | 1.2(4) | 61 |
| 134Cs |        | 6.3(7) | 26(2) |
| 137Cs | 61(2) | 14.1(11) | 72.9(1) – 86.6(1) | 28(3) |
| 232Th | 0.0091(2) | 0.0015(3) | < 4 |
| 228Th | 0.017(2) – 0.069(3) | |
| 233U | < 0.0016 | |
| 227Ac | < (0.2 – 1.1) × 10^{-3} | |
| 238U | 0.0101(2) | 0.009(3) | < 12 |
| 226Ra | 0.022(2) – 0.087(3) | |
Table 9: Radioactive contamination of LiI(Eu), SrI₂(Eu), LaCl₃(Ce), CeBr₃, Cs₂HfCl₆ and YVO₄ crystal scintillators.

| Chain | Source | Activity (mBq/kg) in sample, Reference |
|-------|--------|---------------------------------------|
|       |        | LiI(Eu) | SrI₂(Eu) | LaCl₃(Ce) | CeBr₃ | Cs₂HfCl₆ | YVO₄ |
|       | 26 g  | 49.7 g  | 222 g    | 12 g     | 22 g  |          |      |
| 2 samples | ⊙13 × 11 mm | [261] | [262] | [189] | [150] |          |      |
|        | 40^K  | < 177   | < 200    | < 1.9    | < 0.18 | < 220    |      |
|        | 60^Co | 1.4(4)  | < 19     | < 2.2    |        |          |      |
|        | 82^Br | 18(4)   |          |          |        |          |      |
|        | 90^Sr-90^Y | < 160   | < 90    |          |        |          |      |
|        | 134^Cs| 52(6)   |          |          |        |          |      |
|        | 136^Cs| 25(8)   |          |          |        |          |      |
| 137^Cs | < 22   | 53(11)  | < 0.9    | 830(90)  | < 8.6  |          |      |
|        | 138^La| < 20    | 4.12(8) × 10^5 | 7.4(10)  |        |          |      |
|        | 139^Ce| 4.3(3)  |          |          |        |          |      |
| 137^Sm |        |          |          |          |        | 2.3(5)   |      |
| 152^Eu | < 47   | < 108   |          |          |        |          |      |
| 154^Eu | < 42   | < 67    |          |          |        |          |      |
| 176^Lu | < 42   | < 67    |          |          |        |          |      |
| 181^Hf |        |          |          |          |        | 14.0(7)  | 14.0(7) |
| 232^Th | < 3    | < 88    | < 68     |          |        |          |      |
| 232^Th | < 3    | < 88    | < 68     |          |        |          |      |
| 228^Ac | < 88   | < 68    |          |          |        |          |      |
|        | < 0.4  | 6(2)    | < 0.36   | < 2.0    | < 6.3  | < 10     |      |
| 235^U  | 1.5(4) × 10^4 | < 1.5 | < 26     |          |        |          |      |
| 227^Ac |        | 300(20) |          |          |        |          |      |
| 238^U  | < 40   | < 135   | 1373(2)  |          |        |          |      |
| 234^Th | < 8.6  | 1600(400) | < 26     |          |        |          |      |
| 234m^Pa| < 3.7  | 1200(500) | < 0.5   | < 5(1)   |        |          |      |
| 226^Ra | < 1.1  | 100(14) | < 35     | < 5(1)   |        |          |      |
| 210^Pb | < 480  | < 180   | < 600    |          |        |          |      |
| 210^Po | < 2    | < 60    |          |          |        |          |      |
Table 10: Radioactive contamination of Gd$_2$SiO$_5$ (GSO) and Bi$_4$Ge$_3$O$_{12}$ (BGO) crystal scintillators.

| Chain   | Source | Activity (mBq/kg) in sample, Reference | GSO | BGO |
|---------|--------|----------------------------------------|-----|-----|
|         |        |                                        | 635 g | 1.744 kg | 0.28 – 0.49 kg | 46 g | Table 1 in [263] |
| $^{40}$K | < 14  |                                        | 44   | 204 | 185 | 157 |
| $^{138}$La | < 55  |                                        | 257  | 305 | 290 | 284 |
| $^{147}$Sm | ≈ 100 |                                        | 16  | 22 | 25 | 22 |
| $^{207}$Bi | 520 – 930 | 3000 | 7(2) – 4400 | 232Th | < 6.5 | < 1.1 |
| $^{232}$Th | < 9  | 733 |                                      | 228Pa | < 0.08 |
| $^{228}$Th | 2.287(13) | 107 |                                      | 235U | 0.948(9) |
| $^{212}$Pb | 6(4) |                                      | 208Tl | 3(2) |
| $^{237}$Ac | 0.948(9) |                                      | 231Pa | < 0.08 |
| $^{238}$U | < 2  | 2  | 0.3 |                                      | 230Th | < 9  |
| $^{236}$U | < 2  | 2  | 0.3 |                                      | $^{226}$Ra | 0.271(4) | < 2 |
| $^{210}$Pb | 198 |                                      | 210Po | < 0.8 |
| Total α activity | 40 | 217 |
Table 11: Radioactive contamination of molybdate crystal scintillators.

| Chain | Source | Activity (mBq/kg) in sample, Reference |
|-------|--------|--------------------------------------|
|       |        | Li$_2$MoO$_4$ | Li$_2^{100}$MoO$_4$ | Li$_2$Mg$_2$(MoO$_4$)$_3$ | ZnMoO$_4$ | Zn$^{138}$MoO$_4$ | CaMoO$_4$ | $^{88}$Sr$^{100}$MoO$_4$ |
| $^{40}$K |       | 151 − 242 g. [35] | 186 − 204 g. [35] | 10.2 g. [74] | $\approx$ 340 g. [35] | $\approx$ 380 g. [35] | 82 − 100 g. [229] | $\approx$ 340 g. [35] |
| $^{39}$Sr |       | < 3.2 − 62(2) | < 3.5 | < 1.1 | < 0.018 | < 0.003 | 0.0026(13) | < 23 |
| $^{232}$Th | $^{232}$Th | < 0.018 | < 0.003 | < 0.95 | < 0.0014 | < 0.008 | < 0.7 | < 0.05 |
| $^{232}$Th |       | < 0.018 | < 0.006 | < 1.1 | < 0.005 | < 0.008 | 0.04(2) − 0.42(17) | < 23 |
| $^{238}$U | $^{238}$U | < 0.018 | < 0.005 | < 0.95 | < 0.003 | 0.010(4) − 0.039(7) | < 0.5 | 0.98 |
| $^{238}$U |       | < 0.018 | < 0.007 | < 1.7 | < 0.003 | 0.011(6) − 0.043(10) | < 0.05 | 0.98 |
| $^{230}$Th | $^{230}$Th | < 0.018 | < 0.003 | < 0.95 | < 0.003 | 0.014(3) − 0.023(4) | 0.13(4) − 2.5(5) | 0.065 |
| $^{226}$Ra | $^{226}$Ra | < 0.037 − 0.130(19) | < 0.007 | < 1.7 | < 0.006 | 0.0014(13) − 0.023(4) |
| $^{210}$Pb |       | < 0.018 | < 0.006 | < 0.014 | < 0.017 |
| $^{210}$Pb |       | < 0.018 | < 0.007 | < 1.7 | < 0.017 |
| $^{210}$Pb |       | 0.08(3) − 0.20(4) | 0.06(1) − 0.23(2) | 5.6(13) | 0.575(18) − 1.320(30) | 0.81(3) − 2.39(5) |
| $^{235}$U | $^{235}$U | < 0.018 | < 0.005 | < 0.003 | < 0.003 | < 13 |
| $^{231}$Pa |       | < 0.018 | < 0.003 | < 0.0014 | < 0.008 |
| $^{227}$Th | $^{227}$Th | < 0.018 | < 0.005 | < 0.003 | < 0.009 |
| $^{223}$Ra |       | < 1.1 |
| $^{211}$Bi |       | < 1.1 |

$^{210}$Bi
Table 12: Radioactive contamination of cadmium tungstate (CdWO$_4$) crystal scintillators.

| Chain | Source | Activity (mBq/kg) in sample, Reference |
|-------|--------|---------------------------------------|
|       |        | CdWO$_4$ | $^{106}$CdWO$_4$ | $^{116}$CdWO$_4$ |
|       |        | 72 g     | 1046 g          | 434 g           | 496 g           | 215 g           | 330 g           | 580 g, 582 g |
| $^{40}$K | < 1.4 | < 1.7 | < 5          | < 1.4          | 0.3(1)          | < 0.2          |
| $^{90}$Sr | < 1.4 | < 3   |              |                |                |                |                |
| $^{90}$Sr–$^{90}$Y | < 1 | < 0.3 |              |                |                |                |                |
| $^{110m}$Ag |       |        |              |                |                |                |                |
| $^{113}$Cd | 580 | 580(20) | 558(4)         | 182             | 91(5)           | 100(10)        |
| $^{113m}$Cd | 30(10) | 150(10) | < 3.4         | 116(4) $\times 10^4$ | 1.1(1)         | 460(20)        |
| $^{137}$Cs | < 0.9 | < 0.3 | < 0.3        | < 0.3            |                |                |
| $^{147}$Sm |       |        |              |                |                |                |
| $^{180}$W |       |        |              |                |                | 0.018(7)       |
| $^{190}$Pt | < 0.1 | < 0.1 | < 0.001      | < 0.001         |                |                |
| $^{232}$Th | < 0.026 | < 0.004 | < 0.07 | < 0.07         | 0.053(9)       | < 0.08         |
| $^{228}$Ra | < 0.004 |        |              |                |                |                |
| $^{228}$Th | 0.015(8) | < 0.003 | 0.008(4)   | 0.042(4)       | 0.039(2)       | 0.02 – 0.067(4)$^a$ |
| $^{230}$U | < 0.13 |        |              |                |                |                |
| $^{232}$Ac | < 0.005 | < 0.005 |              |                | 0.0014(2)      | < 0.002        |
| $^{238}$U | 0.038 | < 0.6 | < 0.6       | < 0.6           | 0.3(1) – 0.5(2) |                |
| $^{234}$Pa | < 0.2 |        |              |                |                |                |
| $^{234}$U |        |        |              |                | 0.013          |                |
| $^{230}$Th | < 0.4 | < 0.5 | < 0.5       | < 0.5           |                |                |
| $^{226}$Ra | < 0.04 | < 0.007 | < 0.018 | 0.012(3)   | < 0.004        | < 0.005        |
| $^{210}$Pb |        | < 0.4 |              |                |                |                |
| $^{210}$Po | < 0.063 | < 0.2 |              |                | 0.23(8)        |                |
| Total $\alpha$ activity | 0.26(4) | 2.1(2) | 1.40(10) | 2.10(2) – 2.93(2) |                |                |

$^a$The range of $^{228}$Th activity is due to the decay of $^{228}$Th during the long time measurements.
Table 13: Radioactive contamination of magnesium (MgWO$_4$), calcium (CaWO$_4$), zinc (ZnWO$_4$) and lead (PbWO$_4$) tungstate crystal scintillators.

| Chain | Source | Activity (mBq/kg) in sample, Reference |
|-------|--------|---------------------------------------|
|       |        | MgWO$_4$ | CaWO$_4$ | ZnWO$_4$ | PbWO$_4$ |
| 0.9 g | 54 g   | 189 g | 328 – 740 g | 155 – 310 g | 163 g | 117 – 699 g | 182 g | 454 g$^b$ |
| 210Th | 210Pb  | 313 | 194 | 270 |
| 226Ra | 226Ac  | 16(3) | 0.098(20) | < 0.003 – 0.011(3) | < 0.01 |
|       | 226Th  | 50 | 30 | 0.6(2) | < 1.6 | 0.002(2) – 0.018(2) | < 13 |
| 232Th | 232Th  | 280 | 0.2 | 0.69(10) | < 9 | 0.009(2) | < 0.03 | 0.051(8) |
| 238U | 236U  | 54(4) | 14.0(5) | 38(14) – 330(17) | < 68, 1.01(2) | < 0.01 |
|       | 236Pa  | 400 | 1.08(3) | < 0.007 |
| 234U | 234U  | 1.05(5) | < 0.07 | 0.178(15) |
| 222Ra | 222Ra  | 7(2) | 5.6(5) | 4(2) – 107(11) | < 2.2 – 58, 0.04(1) | < 30 × 10$^3$ | 0.002(1) – 0.025(6) | < 10 | 1.403(43) |
| 214Pb | 214Pb  | 50 | 100 | < 115 |
| 214Bi | 214Bi  | 80 | 0.047(5) | < 159 |
| 210Pb | 210Pb  | 2.4 × 10$^5$ | 900(70) | < 430 | < 190 – 4800(200) | (53 – 79) × 10$^4$ |
| 210Po | 210Po  | 5.7(4) × 10$^5$ | 780(20) | 291(5) | 26(9) – 1316(17) | 0.018(4) | < 0.01 | 186(1) |
| Total $\alpha$ activity | 930$^a$ | 400 | 240(20) – 1400(30) | 1.2 – 107, 3.08(4) | 0.18(3) – 2.3(2) |

$^a$ Estimated from the spectra presented in Fig. 13 of Ref. [268].

$^b$ PbWO$_4$ crystal produced from archaeological lead.
Radioactive contamination of scintillators is summarized in Table 14 where the data for germanium crystals used in ultra-low background HP Ge detector [272, 273] are given for comparison. As one can see in Tables 3-13, authors present their data for different members of the $^{232}$Th and $^{238}$U families. In most cases the measured nuclides are short living daughters of $^{226}$Ra and $^{228}$Th. Therefore, in Table 10 we present data for $^{228}$Th and $^{226}$Ra. Activity of $^{210}$Po is typically measured in scintillators, while its origin is mostly contamination by lead ($^{210}$Pb). For this reason data on $^{210}$Pb activity is presented in Table 14. However, one should keep in mind that equilibrium between $^{210}$Pb and its daughter $^{210}$Po is broken as usual, as it was discussed above.
Table 14: Radioactive contamination of scintillators (mBq/kg). Data for germanium crystals of HP Ge detectors [272, 273] are given for comparison.

| Scintillator | $^{86}$K | $^{232}$Th | $^{228}$Ra | $^{210}$Pb | Total α (U + Th) | Particular radioactivity |
|--------------|---------|-----------|-----------|-----------|-----------------|-------------------------|
| Plastic scintillator | 2 | $< 0.0001 - 9$ | 0.09 - 1 | $< 2 \times 10^{-6}$ - 0.06 | 14C | 14C |
| Liquid scintillator | $< 1 \times 10^{-3} - 3 \times 10^{-7}$ | $< 4 \times 10^{-9} - 3 \times 10^{-7}$ | 3 | $< 2 \times 10^{-6}$ - 0.06 | 14C | 14C |
| LaF₃(W) | $< 5$ | $< 0.6$ | 3 | | | 186W |
| CaF₂(Eu) | $< 7$ | 0.1 - 41 | 0.03 - 75 | 0.9 | 8 | 42Ca, 131Eu |
| SrF₂ | 21 | 196 | 72² | | | |
| CeF₃ | $< 330$ | 1010 | $< 60$ | $< 280$ | | |
| BaF₂ | $1.4 \times 10^{3}$ | ($1.4 - 7.8) \times 10^{3}$ | 990 | | | |
| YAG(Nd) | | | | | | |
| Li₄Eu(BO₃)₃ | $< 1500$ | 3.5⁴ | 2.9 | 0.2 | 42Eu, 131Eu, 134Eu | 42Eu, 131Eu, 134Eu |
| α | $< 0.0004 - 0.02$ | $< 0.0004 - 0.03$ | $< 0.1$ | | 65Zn, 75Se, 82Se |
| β | $0.02 - 0.03$ | $0.02 - 0.04$ | 0.1 - 0.3 | | | 82Se |
| LaCl₃(Ce) | $< 0.4$ | $< 40$ | $< 20$ | | 134La, 238U | 134La, 238U |
| Cs₂HCl₆ | $< 0.18$ | $< 6$ | $< 0.02$ | | | |
| Li(II) | $< 180$ | $< 0.4$ | $< 1.1$ | | | 176Y |
| Na(II) | 0.3 - 81 | 0.0068 - 0.18 | $< 0.0002 - 1$ | 0.02 - 10 | 131I | 131I |
| Sr(II) | $< 200$ | 60 | $< 180$ | | | 132Sn |
| Ca(II) | 17 | $< 0.4$ | $< 1.2$ | | | 133Cs, 134Cs |
| Cs(II) | 17 | 0.0009 - 0.07 | 0.0009 - 0.09 | | | 134Cs, 137Cs |
| Ce(II) | 1.9 | $< 2$ | $< 0.5$ | $< 600$ | | 82Br, 136Cs |
| GSO(Ce) | 14 | 2.3 - 107 | 0.27 | 200 | 40 - 220 | 137Cs, 137Cs |
| BGO | 7 | 6 | | | | 137Cs, 137Cs |
| Li₂MoO₄ | $< 3 - 60$ | 0.018 | $< 0.04 - 0.13$ | (0.08 - 0.2)² | ²⁶⁰Mo | ²⁶⁰Mo |
| Li₂MoO₄ | $< 3$ | $< 0.006$ | $< 0.007$ | (0.06 - 0.23)² | ²⁶⁰Mo | ²⁶⁰Mo |
| Li₂MoO₄ | $< 1.1$ | 1.2 | $< 2$ | | ²⁶⁰Mo | ²⁶⁰Mo |
| ZnMoO₄ | $< 0.005$ | $< 0.006$ | 0.6 - 1.3² | | | ²⁶⁰Mo |
| ZnMoO₄ | $< 0.008$ | 0.014 - 0.023 | 0.8 - 2.4² | | | ²⁶⁰Mo |
| CaMoO₄ | $< 1.1$ | 0.04 - 0.1 | 0.13 - 2.5 | $< 8 - 550$ | | ⁴⁴Ca, ⁴⁴Mo |
| CaMoO₄ | $< 1.1$ | 0.04 - 0.1 | 0.13 - 2.5 | $< 8 - 550$ | | ⁴⁴Ca, ⁴⁴Mo |
| MgWO₄ | $< 1.6 \times 10^{3}$ | 50 | 50 | 5.7 $\times 10^{6}$ | 5.7 $\times 10^{6}$ | ²⁶⁶W |
| CaWO₄ | $< 12$ | 0.6 | 0.04 - 107 | $< 190 - 480$ | 1 - 1400 | ⁵⁴Ca, ²⁶⁶W |
| ZnWO₄ | $< 0.02$ | 0.002 - 0.018 | 0.002 - 0.025 | $< 0.01$ | 0.18 - 2.3 | ²⁶⁷Sr, ⁵⁴Zn, ²⁶⁶W |
| CaWO₄ | $< 1.7 - 4$ | $< 0.003 - 0.015$ | $< 0.007$ | $< 0.06$ | 0.26 | ¹¹⁰Cd, ¹⁴⁴Cd, ¹⁴⁶W |
| ¹⁴⁴CaWO₄ | $< 1.4$ | 0.04² | 0.1² | $< 0.2$ | 2.1 | ¹¹⁰Cd, ¹⁴⁴Cd, ¹⁴⁶W |
| ¹⁴⁴CaWO₄ | $< 0.2 - 0.3$ | 0.02 - 0.07 | $< 0.004$ | 0.23 | 1.4 - 2.9 | ¹¹⁰Cd, ¹⁴⁴Cd, ¹⁴⁶W |
| PbWO₄ | $< 13$ | $< 10$ | (5 - 8) $\times 10^{6}$ | | | ²¹⁰Po, ²¹⁴Po |
| PbWO₄ | $< 0.05$ a | 1.4 | 190 | | | ²¹⁰Po, ²¹⁴Po |
| HP Ge | $< 2 \times 10^{-6}$ | $< 2 \times 10^{-6}$ | | | | ²⁹⁳Ge |

* Activity of $^{212}$Th.

² Activity of $^{238}$U.

³ Activity of $^{210}$Po.

⁴ Activity of $^{238}$U + $^{226}$Ra.

⁵ Produced from ancient lead.
Liquid scintillators are the most radiopure scintillation materials with a n Bq/kg – pBq/kg radiopurity level of \(^{40}\)K, U and Th \[^{237, 238}\]. However, cosmogenic \(^{14}\)C remains the main source of the background counting rate (0.4 – 0.7 mBq/kg, at energies below 0.25 MeV) of large low-background liquid scintillation detectors, despite the significant efforts to reduce its concentration. Radioactive contamination of plastic scintillators is significantly higher. It can be explained by polymerization process and mechanical treatment of the material, as well as by absence of a strong motivation to obtain highly radiopure scintillation material similar to the Borexino and KamLAND experiments. Crystal scintillators like ZnWO\(_4\), CdWO\(_4\) (including produced from enriched \(^{106}\)Cd and \(^{116}\)Cd), Li\(_2\)MoO\(_4\), ZnMoO\(_4\) (including enriched in \(^{100}\)Mo), ZnSe (including enriched in \(^{82}\)Se), specially developed for low background experiments CaF\(_2\)(Eu), NaI(Tl), CsI(Tl) and CaMoO\(_4\) have rather low contamination by \(^{226}\)Ra and \(^{228}\)Th on the level of \(~0.001 – 0.1\) mBq/kg.

A level of crystal scintillators radiopurity is determined first of all by their chemical composition. For instance, CdWO\(_4\) and ZnWO\(_4\) crystals always show low level of internal activity, while CaWO\(_4\) has much higher level of radioactive contamination. Fig. [12] demonstrates the large difference in radiopurity of CaWO\(_4\), CdWO\(_4\) and PbWO\(_4\). Scintillators containing rare earth elements (GSO, CeF\(_3\)) have much higher level of radioactive trace pollution too. It is due to source of rare earth mining: they usually are extracted from monazites – minerals containing a few percents of uranium and thorium. Energy spectra of CaF\(_2\)(Eu) and CeF\(_3\) crystal scintillators measured in the same conditions of the DAMA R&D low background set-up in the Gran Sasso underground laboratory of I.N.F.N. (Italy) are presented in Fig. [13]. The background counting rate of the CeF\(_3\) detector is two orders of magnitude higher due to the higher contamination by thorium and uranium. Rather high radioactive contamination of BaF\(_2\) by radium (\(^{226}\)Ra and \(^{228}\)Ra) can be explained by similar chemical properties of barium and radium. It should be stressed that the chemical and physical processes involved in scintillators production are sensitive to the chemical properties of materials. Therefore, it might be more accurate to discuss contamination of materials by chemical elements, not by certain isotopes.

Presence of radioactive elements (see Table 2) obviously determines radioactivity of scintillators like \(\beta\) active \(^{113}\)Cd in CdWO\(_4\), \(\alpha\) active \(^{152}\)Gd in GSO, \(^{138}\)La in LaCl\(_3\) and LaBr\(_3\), \(^{176}\)Lu in Lu\(_2\)SiO\(_5\) and LuI\(_3\). Beta active \(^{210}\)Pb is usually present in PbWO\(_4\) and PbMoO\(_4\) (however, this problem can be overcome by producing lead containing scintillators from archaeological lead \[^{51, 216, 161}\]).

Two neutrino 2\(\beta\) decay, despite it is the rarest decay ever observed, becomes one of the most significant background sources in the experiments aiming to search for 0\(\nu\)2\(\beta\) decay with smaller \(Q_{2\beta}\). For this reason the AMoRE collaboration developed CaMoO\(_4\) crystal scintillators enriched in the isotope \(^{100}\)Mo and depleted in \(^{48}\)Ca \[^{274, 56}\]. Another example is CdMoO\(_4\) crystal scintillators proposed to search for 0\(\nu\)2\(\beta\) decay of \(^{116}\)Cd and \(^{100}\)Mo \[^{275}\]: in this case the 2\(\nu\)2\(\beta^-\) decays of \(^{100}\)Mo will generate background in the region of interest of \(^{116}\)Cd.

5 Development of radiopure scintillation materials

A strong R&D is required to obtain a radiopure scintillation material. There were several systematic programmes to elaborate radiopure scintillators. Extremely radiopure liquid scintillators were developed for the Borexino \[^{276}\] and KamLAND \[^{277}\] neutrino experiments. Two factors which determine success of the projects are as following: 1) organic materials in
Figure 12: Energy spectra of CaWO$_4$ (189 g, 1734 h), CdWO$_4$ (448 g, 37 h), and PbWO$_4$ (185 g, 2.15 h) scintillation crystals measured in the low background set-up in the Solotvina Underground Laboratory (the PbWO$_4$ crystal was measured without shield). The CaWO$_4$ crystal is considerably polluted by radionuclides from U and Th chains [230]. Beta decay of $^{113}$Cd ($T_{1/2} = 8.04 \times 10^{15}$ years) dominates in the low energy part of the CdWO$_4$ spectrum [146, 147]. PbWO$_4$ crystal is contaminated by $^{210}$Pb [271].
Figure 13: Energy spectra of CeF$_3$ (49 g, 2142 h) and CaF$_2$(Eu) (370 g, 7426 h) scintillation crystals measured in the DAMA R&D low background set-up in the Gran Sasso underground laboratory of I.N.F.N. (Italy). The CeF$_3$ crystal is considerably polluted by radionuclides from Th and U chains on the $\sim$ Bq/kg level (see for details [205]). The background of the CaF$_2$(Eu) detector is two orders of magnitude lower, and caused by $\alpha$ activity of U/Th daughters on the level of a few mBq/kg [159]. In the low energy part of the spectrum accumulated with the CaF$_2$(Eu) crystal one can see peaks from cosmogenic (or / and neutron induced) $^{152}$Eu ($\approx$ 10 mBq/kg), as well as $\alpha$ peak of $^{147}$Sm presented in the crystal with an activity of $\approx$ 0.3 mBq/kg.
principle are much less contaminated in comparison to inorganic materials; 2) liquids can be effectively purified by distillation, nitrogen purging, water extraction and/or column purification [276, 278].

Very low background NaI(Tl) scintillators have been produced by Saint-Gobain for the DAMA/LIBRA dark matter experiment [209, 279, 280]; several R&D are in progress to develop radiopure NaI(Tl) scintillators [213, 252, 253, 256, 257]. Low background CsI(Tl) were developed in the framework of the KIMS dark matter experiment [187, 208]. Dependence of radiopurity of CsI(Tl) on crystal growth conditions was studied in [208]. Authors reported a clear increase of $^{226}$Ra and $^{228}$Th activity along the crystals during the crystal growth process due to increase of the melt contamination. Clear increase of radioactive contamination along the crystal boule was observed in $^{116}$CdWO$_4$ crystal [281].

Achievement of a high radiopurity level of scintillators needs a variety of special measures as it was demonstrated by development of radiopure crystal scintillators for double beta decay experiments: $^{106}$CdWO$_4$ [266], $^{116}$CdWO$_4$ [193], $^{48}$Se$_{\text{depl}}$Ca$^{100}$MoO$_4$ [37, 282], Zn$^{82}$Se [283], Zn$^{100}$MoO$_4$ [284, 285, 286, 287, 35], and Li$^{210}$MoO$_4$ [62, 288, 35].

A programme to develop radiopure scintillation materials for experiments to search for rare processes could comprise the following steps [289]:

1. Careful selection and deep purification of raw materials is supposed to be the most important issue that needs addressing. Purification of metals (Zn, Cd, Pb, Bi) by vacuum distillation, zone melting, and filtering are very promising approaches [290, 291, 292], while further study is necessary for the purification of Li, Na, Ca, Se, Cs, Ba, Ce, Gd, W, Ti. Molybdenum can be deeply purified by combination of a double sublimation of molybdenum oxide with subsequent recrystallization in aqueous solutions [285].

2. Two step re-crystallization, involving inspection and assessment of the produced scintillators after each step. A promising result was obtained with the $^{116}$CdWO$_4$ crystal scintillator by re-crystallization. The radioactive contamination of a sample of $^{116}$CdWO$_4$ scintillator by thorium was reduced by one order of magnitude after the second crystallization procedure [281]. A larger number of recrystallization steps is limited by evaporation of components of melt (e.g. dominant evaporation of cadmium from CdWO$_4$ melt, or molybdenum from Li$_2$MoO$_4$, CaMoO$_4$, ZnMoO$_4$), which leads to violation of stoichiometry of the melt. Besides, one could expect some influence of growing process to radioactive contamination of crystal scintillators, which effects were never systematically studied. One could expect that at a certain stage of such an R&D, development of special low background growth facilities will be necessary. An interesting R&D was performed in [252] aiming at development of highly radiopure NaI(Tl) crystal scintillators. Authors observed a significant improvement of the crystal radiopurity level by utilization of high purity graphite crucible. Investigations of ceramics used in the growing set-ups was performed in the framework of radiopure ZnWO$_4$ crystal scintillators R&D [150]. We would like to stress that the low-thermal gradient Czochralski crystal growth method provides high quality large volume radiopure crystal scintillators [293]. The technique is especially suitable for enriched crystal scintillators production [294].

3. Screening at all stages through ultra-low background $\gamma$-spectrometry is needed in the production of compounds for crystal growing (choice of raw materials, quality control of purified elements and compounds).

4. All work should be done using highly pure reagents, lab-ware and water. Careful protection from radon should be foreseen (especially in the case of scintillators R&D for dark matter experiments). All chemistry should be done in clean room conditions, and, as far as possible,
in radon free atmosphere.

5. Cosmogenic activation is expected to be one of the most significant sources of background as the low counting technique improves. Therefore, underground facilities for crystal scintillators production could be a natural step to obtain highly radiopure scintillators.

6 Conclusions

Scintillation detectors are widely used to search for rare processes in nuclear and astroparticle physics. Radioactive contamination of scintillation materials can be measured by indirect (inductively coupled plasma mass spectrometry and neutron activation are the most sensitive approaches) and direct (first of all, by low-background HP Ge $\gamma$ spectrometry) methods. However, the most sensitive method to measure internal contamination of a scintillator are low background measurements when the scintillator operates as a detector. Time-amplitude analysis allows to detect at the $\mu$Bq/kg level the fast sub-chains of the $^{232}$Th, $^{235}$U and $^{238}$U families which are in equilibrium with $^{226}$Th (from the $^{232}$Th family), $^{226}$Ra ($^{238}$U) and $^{227}$Ac ($^{235}$U). Alpha active nuclides can be determined by using pulse-shape discrimination technique. Estimation of $\beta$ active nuclides can be done by fit of measured energy spectra using Monte Carlo simulated models of background.

Liquid scintillators are the most radiopure scintillation materials with the best up to date achieved residual radioactive contamination level of nBq/kg – pBq/kg. Radioactive contamination of crystal scintillators varies in a wide range. The most radiopure crystal scintillators are ZnWO$_4$, CdWO$_4$, Li$_2$MoO$_4$, ZnMoO$_4$, ZnSe and specially developed for dark matter experiments NaI(Tl) and CsI(Tl) those radioactive contamination by $^{228}$Th and $^{226}$Ra does not exceed the level of $\sim 0.01$ mBq/kg. Main sources of internal radioactivity of scintillators are daughters of U/Th families, $^{40}$K, radioactive isotopes of elements which are part of a scintillator composition. Scintillation materials containing rare earth elements have comparatively high level of U/Th contamination. Equilibrium of the $^{232}$Th, $^{235}$U and $^{238}$U chains is usually broken in scintillators.

Next generation $2\beta$ and dark matter experiments call for extremely low ($\sim 0.1 – 1\mu$Bq/kg) level of radioactive contamination of crystal scintillators, that can be obtained in the framework of special programmes that should include deep purification of raw materials, careful screening at all the stages, application of double crystallization. The programmes should involve use of radiopure reagents, lab-ware, equipment and installations, production of raw materials, crystal growing and their storage in radon free atmosphere. Special efforts are necessary to prevent cosmogenic and neutron activation of materials. Development of special underground low background growth facilities may be necessary to reach a further progress in production of radiopure crystal scintillators. Tests of crystal scintillators radioactive contamination on the 0.1 – 1 $\mu$Bq/kg level can be realized by ultra-low background direct measurements when a scintillator serves as a detector of its internal radioactivity. The technique of cryogenic scintillating bolometers provides the highest sensitivity to $\alpha$ active contaminations of crystal scintillators.

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