Depth-profile study of $^{210}$Pb in the surface of an NaI(Tl) crystal

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Abstract

The surface $^{210}$Pb is one of the main background sources for dark-matter-search experiments using NaI(Tl) crystals, and its spectral features associated with the beta-decay events for energies less than 60 keV depends on the depth distribution of $^{210}$Pb in the surface of an NaI(Tl) crystal. Therefore, we must understand the profile of surface $^{210}$Pb to precisely model the background measurement in the low-energy region for the low-background experiment using NaI(Tl) crystals. We estimate the depth profile of the surface $^{210}$Pb contamination by modeling the measured spectrum of the alpha emission from the decay of $^{210}$Po at the decay sequence of the surface $^{210}$Pb contamination that is obtained using an $^{222}$Rn-contaminated crystal. Moreover, we describe the energy spectra of the surface contaminations by performing Geant4 Monte Carlo simulations that are used for fitting an exponential curve, which is a function of the surface depth, with the measured data, by using the log likelihood method. The low- and high-energy events from the beta decay of surface $^{210}$Pb are also modeled to improve the depth profile for shallow depths. We simulate the energy spectra from beta decays of $^{210}$Pb that are exponentially distributed in the surface by following two exponential functions that consider mean-depth parameters in the data fitting; we observed that the energy spectra are in good agreement with the measured data.

Keywords: Surface contamination, depth profile, $^{210}$Pb, $^{210}$Po, NaI(Tl) crystal

1. Introduction

The presence of dark-matter particles in the Universe has been evidenced via numerous astronomical observations [1,2]. Weakly interacting massive particles (WIMPs) are one of the most attractive candidates for being dark-matter particles [3,4]. Many experiments have been conducted for directly searching WIMPs in our galaxy by looking for nuclear recoils that are produced by WIMP–nucleus scattering [5,6]; however, null results have been reported thus far. However, a notable exception is the DAMA/LIBRA experiment, which has consistently reported the observation of an annual event-rate modulation in an array of NaI(Tl) crystal detectors; this modulation could be interpreted as a dark-matter signal [7]. Furthermore, there are several NaI(Tl)-crystal-based experiments [8,9,10] to test DAMA/LIBRA’s observation of an annual event-rate modulation with the statistical significance of more than 12.9 $\sigma$ [14,15].

Searching an annual modulation signal requires the complete understanding of background sources and complete simulation that accurately models the background-energy spectra measured using the detector; therefore, a background model, which is based on Monte Carlo simulations, has been built using the Geant4 toolkit [16,17,18,19,20,21,22]. In the model, it has been reported that the low-energy contribution from the beta-decay of $^{210}$Pb in the surface of NaI(Tl) crystals is one of the dominant background sources; furthermore, it has been suggested that sources are attributed to the $^{222}$Rn contamination that occurred anytime during the powder- and/or crystal-processing stages [17,18].

As depicted in Fig. 1, the short-lived $^{222}$Rn decays to $^{218}$Po, which can be deposited on the crystal surface as a reactive metal and recoils $^{214}$Pb into the crystal surface during its subsequent alpha decay [23,24]. The $^{214}$Po alpha decay after the beta decays of $^{214}$Pb results in the implantation of $^{210}$Pb deeper into the surface; consequently, $^{210}$Pb has an implanted distribution in the surface and also a long half-life ($t_{1/2} = 22.3$ years), thereby acting as a background source for the low-energy region in an NaI(Tl) crystal; in addition, it contributes to the low-energy spectra by the beta decays to $^{210}$Po, which, subsequently, decays via alpha emission to $^{206}$Pb.

Because the beta decay to $^{210}$Bi results in low-energy events via the emissions of electrons and $\gamma$/X-ray, the spectral features of these events for energies less than...
60 keV depend on the depth distribution of $^{210}$Pb within the crystal surface. Therefore, we must completely understand the surface $^{210}$Pb profile to precisely model the background measurement in the low-energy region for the low-background experiment using NaI(Tl) crystals. Additionally, there are other experiments that improved the low-background experiment using NaI(Tl) crystals. Additionally, there are other experiments that improved the techniques to reject the surface background, as reported in Refs. [25, 26, 27].

To estimate the depth profile of the surface $^{210}$Pb contamination, we modeled the measured spectrum due to the alpha emission from the decay of surface $^{210}$Po ($t_{1/2} = 138$ days) at the decay sequence of surface $^{210}$Pb ($t_{1/2} = 22.3$ years), by performing Monte Carlo simulations using the Geant4 version 10.4.p02 (Sect. [3]). In addition, the low- and high-energy events from the beta decay of surface $^{210}$Pb are modeled to improve the depth profile for shallow depths (see Sect. [4]).

2. Experimental setup

A copper encapsulated cylindrical NaI(Tl) crystal - with 8 cm diameter, 10 cm length, and 1.28 kg mass - was used in the experiment. It was made using the same ingot as the two crystals (C6 and C7) employed in the COSINE-100 experiment [23, 24]. The crystal was cut into two pieces, and the surface of one piece (Crystal B) was exposed to $^{222}$Rn gas from a $^{222}$Rn source for two weeks, while the other piece (Crystal A) was retained as clean. Subsequently, both the pieces were attached facing each other by inserting a 4-$\mu$m thick aluminized mylar film between them; the film allowed only alpha and beta particles to pass through it, and not photons. In addition, two photomultiplier tubes (PMTs), which detected the scintillation signals coming from the crystals, were attached at the ends of both the crystal. The signal was saved only when the height of the pulse in both the PMTs was greater than the preset threshold. Figure 2 depicts the crystal–PMT detector module, which was installed inside the CsI(Tl) crystal array setup that had been previously deployed in the KIMS experiment in Yangyang underground laboratory [30]. Furthermore, Nitrogen gas was supplied into the detector setup to avoid radon contamination and maintain a stable humidity level; the details of the experimental setup and data-recording conditions are described in Ref. [31].

To minimize the contributions from the decay of the mother isotopes of $^{210}$Pb and to ensure events primarily from the $^{210}$Pb decay, only the data recorded after 70 days from the date of detector installation were used in this study; notably, the amount of data corresponds to 110 days. The data were prepared using the following criteria. An event that has hits only in the NaI(Tl) crystals (Crystals A and B) is called a single-hit event, whereas the one that has accompanying hits in the surrounding CsI(Tl) crystals too is called a multiple-hit event. For this study, we selected single-hit events since multiple-hit events are mostly due to external background sources. In addition, a timing coincidence within 200 ns was required between Crystals A and B, called coincidence events, to consider the events that were mainly induced by the deposition on the crystal surface. Furthermore, the energy scale below 100 keV was calibrated using a $\gamma$-ray source and the background spectra from several radioactivities of each crystal. For Crystal A, we used the peaks at 12, 28, and 46.5 keV from the surface $^{210}$Pb, and at 59.54 keV from a $^{241}$Am $\gamma$-ray source. The peak at 76.63 keV from the $^{214}$Bi decay was used for Crystal B. Above 100 keV, energy calibration was performed using peaks at 609 and 1120 keV from $^{210}$Bi for both the NaI(Tl) crystals.
3. Modeling alpha spectra of $^{210}$Po decay in the crystal surface

3.1. Simulations of the surface $^{210}$Po contamination

As described in Sect. 2, the surface of Crystal B was exposed to $^{222}$Rn gas from a $^{226}$Ra source for two weeks; therefore, $^{210}$Pb at the decay sequence of $^{222}$Rn exhibits an implanted distribution in the surface with a long half-life ($t_{1/2} = 22.3$ years); it results in the low-energy spectra because of the beta decays to $^{210}$Po, which subsequently decays via alpha emission to $^{206}$Pb. To estimate the depth profile of the surface $^{210}$Pb contamination, we modeled the measured spectrum using the clean crystal, i.e., Crystal A; the spectrum is attributed to the alpha emission from the decay of surface $^{210}$Po ($t_{1/2} = 138$ days) of the contaminated Crystal B, by simulating all the decay chains of $^{210}$Pb by generating it in the surface of Crystal B. We, thus, generated $^{210}$Pb using different depths of the surface of Crystal B, and investigated the correlation of alpha spectra measured using clean Crystal A in terms of the surface depth. Because of both the thin mylar layer and the small energy depositions on the contaminated crystal, i.e., Crystal B, the measured energy of the alpha particles was below the full energy deposition of $^{210}$Po, as reported in Ref. [31]; in addition, their small energy depositions on Crystal B vary depending on the depth of the surface, thereby resulting in different alpha spectra on clean Crystal A, as depicted in Fig. 3. These were used as inputs when fitting the simulations based on an exponential curve, which was a function of depth of the surface, with the measured data; the details are described in Sect. 3.2.

To compare the energy spectrum of the alpha particles from the simulations with the data, we used a light yield ratio of $\alpha/\beta$ as a function of the energy of the alpha particle, as determined using the following two peak positions: one is of a Q-value of 5.407 MeV from the alpha decay of $^{210}$Po and the other is of the alpha particle with 5.304 MeV that deposits the average energy of 3.0 MeV in Crystal B. This ensures maximal stopping energy loss at the end of its track when passing through the crystal with inactive region on the surface. Upon contaminating a crystal by exposing it to $^{222}$Rn gas, a foggy layer was formed on its surface; this layer can affect the scintillation performance by acting as an inactive region or a dead layer (DL) [32]; therefore, the contaminated crystal, i.e., Crystal B, appeared obscured, while the clean Crystal A appeared transparent. We, thus, considered a DL in the surface of contaminated Crystal B.

3.2. Results and comparison with measured data

To model the surface $^{210}$Pb spectrum, we generated $^{210}$Pb decays at random locations within the surface thickness of 10 $\mu$m in the contaminated Crystal B, which was divided into 100 bins, each with a thickness of 0.1 $\mu$m, to control the fraction of events on the basis of depth. We assume that they are exponentially distributed in the surface, as follows:

$$p_0 \cdot \exp\left(-\frac{x_d}{p_1}\right) \cdot E_d$$  \hspace{1cm} (1)

where $x_d$ denotes the depth in $\mu$m, $E_d$ the energy spectrum of the events within the depth, and $p_0$ and $p_1$ the amplitude and mean depth of the exponential distribution, respectively. Subsequently, we fitted the alpha spectrum measured using the clean Crystal A by employing the log likelihood method, which allows the simulated spectrum to follow Eq. (1), resulting in the best-fit parameters of $p_0$ and $p_1$. In the fit, we also included five DLSs of various thicknesses from 1 to 5 $\mu$m as follows: 0~1, 0~2, 0~3, 0~4, and 0~5 $\mu$m, and their fractions were treated as free-floating parameters. Figure 4 depicts the fitted result (solid red
line) for the alpha spectrum on the clean Crystal A; the result is in good agreement with the data (filled black circles). It provides the depth distribution of $^{210}\text{Po}$ following the exponential function with the mean-depth parameter, $p_1 = (0.88 \pm 0.03) \mu\text{m}$, as the best-fit result. It also shows that the alpha-decay events from $^{210}\text{Po}$ generated on the surface, for the DLs with thicknesses of 1, 2, and 3 $\mu\text{m}$, are considered one of the dominant contributors.

4. Modeling the energy spectra of $^{210}\text{Pb}$ beta decay within the crystal surface

The beta decays to $^{210}\text{Bi}$ from $^{210}\text{Pb}$ produced low-energy events via the emissions of electrons and $\gamma$/X-ray; therefore, their spectral features for energies less than 60 keV depend on the depth distribution of $^{210}\text{Pb}$ on the crystal surface. To understand the energy spectra from the beta decays of $^{210}\text{Pb}$, we simulated them by generating $^{210}\text{Pb}$ at random locations within the surface thickness of 1 $\mu\text{m}$ in Crystal B. The simulated spectra for both the contaminated Crystal B and clean Crystal A are depicted in Fig. 5 (a) and (b), where each color represents the beta decays of $^{210}\text{Pb}$ (dotted red line) and $^{210}\text{Bi}$ (dashed blue line), respectively. The peaks at approximately 10 and 46 keV are attributed to the X-rays and 46.5 keV $\gamma$-ray from the decays of $^{210}\text{Pb}$. In addition, the conversion electrons contribute to the peaks at approximately 20 and 35 keV. Therefore, these spectral features depend on the beta-decay events distributed within a shallow surface depth.

To verify the depth profile derived using the alpha-spectrum model, which is described in Sect. 3.2, we compared the low-energy spectra measured using each of Crystals A and B with the simulated spectra of $^{210}\text{Pb}$ that is distributed in the surface of Crystal B, in accordance with the depth profile derived using the alpha spectrum; we observed that the low-energy spectra were not satisfactorily reproduced using the simulated spectra in the low-energy region, as depicted in Fig. 6.

If the $^{210}\text{Po}$ contamination is zero at the instance when the initial $^{210}\text{Pb}$ contamination occurred, it will grow with the $^{210}\text{Po}$ half-life, i.e., $\tau_{^{210}\text{Po}} = 138$ days, until reaching an equilibrium. This change in the total alpha rate with time

Figure 5: Low-energy spectra due to the beta decay of $^{210}\text{Pb}$ to $^{210}\text{Po}$: in the contaminated Crystal B (a), and in the clean Crystal A (b).

Figure 6: Low-energy spectra due to the beta decay of $^{210}\text{Pb}$ generated in the surface with the mean-depth parameter, $p_1$, include all the energy deposited in the clean Crystal A (bottom) and in the contaminated Crystal B (top). The energy spectra are compared with the measurement results (filled black circles).
can provide information regarding the total amount of the surface $^{210}$Pb contamination. Because the 110 day data used in this study were recorded after 70 days from the time when the initial $^{210}$Pb contamination occurred and when an equilibrium was not yet reached, the $^{210}$Po contamination is lower than the surface $^{210}$Pb contamination. However, the spectral feature of the simulation is not satisfactorily matched to the data, although it is scaled. The spectral-feature discrepancy between the data and simulation can be explained as follows: most alpha events occurring at the shallow depth of less than 1 $\mu$m are not tagged as coincidence events when it is dominated by an inactive region and when the path length of the recoiled $^{206}$Pb with 106 keV kinetic energy is as small as $\sim$50 nm; therefore, it results in the lack of full understanding of the depth profile for shallow depths. Therefore, to improve the model for better understanding the depth profile of $^{210}$Pb for shallow depths, we assume that they are exponentially distributed in the surface of Crystal B, via following two exponential functions:

$$p_0 \cdot \exp\left(-\frac{x_d}{p_1}\right) + p_2 \cdot \exp\left(-\frac{x_d}{p_3}\right) \cdot E_d \quad (2)$$

where we constrain the parameter $p_1$ using $(0.88 \pm 0.03) \mu$m and treated $p_0$, $p_2$, and $p_3$ as free-floating parameters in the data fitting, provided that the inactive region was derived using the alpha-spectrum model.
In the fit, the low- and high-energy spectra of the beta-decay events, as well as the energy deposited in each of Crystals A and B, are simultaneously fitted using simulations. The fitted energy spectra for both Crystals A and B (solid red line) are compared with the measurement results (filled black circles), as depicted in Fig. 8. The overall energy spectra (solid red lines) for both the clean Crystal A and contaminated Crystal B are in good agreement with the data, not only for low-energy events but also for high-energy events. The high-energy events are dominantly the result of the external 226Ra from the PMTs that also slightly contributes to the low-energy region and beta decay from 210Bi to 210Po in the crystal surface that ends with 1.2 MeV. As a result of the fit, the mean of the exponential depth distribution with p1 is as shallow as (0.13±0.08) μm. The ratio of the amplitudes of two exponential distributions, p0 to p1, is approximately 0.1; therefore, the low-energy events are primarily attributed to the beta decays of 210Pb at a given location, following the exponential function with the mean depth of 0.13 μm. In the fit, we included four DLs having various thicknesses from 1 to 4 μm, and it is in good agreement with the data when it is dominated by a 3-μm thick DL.

According to the decay chain for 210Pb, most events with energy deposition in the crystals are attributed to the conversion electrons, Auger electrons, and γ/X-rays, followed by the beta electrons from the decay to 210Bi of 210Pb in the surface of Crystal B. Therefore, the coincidence events might possess an energy correlation between Crystals A and B. In Fig. 8, we depict a scatter plot of the energy deposition in Crystal A versus that in Crystal B. Figure 8(a) and (b) depict the simulated results both without including a DL and upon including a DL, respectively; Fig. 8(c) depicts the measured data. In the simulation, we included a 3-μm thick DL while generating 210Pb decays at random locations within the surface thickness of 0.5 μm in the contaminated Crystal B; subsequently, we reproduced the distinct islands at approximately 20 and 30 keV in one crystal and at 30 and 20 keV in the other one, as evident from the measured data, while the islands are not depicted in Fig. 8(a), which does not consider a DL.

5. Conclusion

We have studied the depth profile of the surface 210Pb contamination of an NaI(Tl) crystal by modeling the measured energy spectra with the simulated spectra using the Geant4 toolkit. We first fitted the alpha-decay events with the simulated spectrum from the alpha decay of 210Po that is exponentially distributed within the crystal surface by following the exponential function, under the condition of an inactive region; subsequently, we showed that the fitted result was in good agreement with the alpha data. In addition, to improve the depth profile for shallow depths below 1 μm, we studied the energy spectrum from the beta decay of 210Pb whose spectral features were attributed to γ/X-rays and conversion electrons that were mainly distributed within the shallow depth. Subsequently, we showed that all the fitted results of both the clean and contaminated crystals were in good agreement with the data for both the low- and high-energy events.

Using this study, we can provide the quantitative understanding of the depth profile of the surface 210Pb contamination in an NaI(Tl) crystal. Furthermore, our analysis method and results can be applied to NaI(Tl) experiments, in general, to describe the backgrounds in case of 210Pb surface contamination.

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References

[1] E. Komatsu et al. (WMAP Collaboration), Astrophys. J. Suppl. 192 (2011) 18.
[2] P. A. R. Ade et al. (Planck Collaboration), arXiv:1303.5076.
[3] B. W. Lee and S. Weinberg, Phys. Rev. Lett. 39 (1977) 165.
[4] G. Jungman, A. Kamionkowski, G. Griest, Phys. Rep. 267 (1996) 195.
[5] R. Gaitskell, Annu. Rev. Nucl. Part. Sci. 54 (2004) 315.
[6] L. Baudis, Phys. Dark Univ. 1 (2012) 94.
[7] R. Bernabei, DAMA collaboration, Eur. Phys. J. C 56 (2008) 333; R. Bernabei et al. (DAMA Collaboration), Eur. Phys. J. C 67 (2010) 39; R. Bernabei et al. (DAMA Collaboration), Eur. Phys. J. C 73 (2013) 2648.
[8] G. Adhikari, et al. (COSINE-100 collaboration), Search for a dark matter-induced annual modulation signal in NaI(Tl) with the COSINE-100 experiment, Phys. Rev. Lett. 123, 031302 (2019).
[9] E. Barbosa de Souza, et al., First search for a dark matter annual modulation signal with NaI(Tl) in the Southern Hemisphere by DM-Ice17, Phys. Rev. D 95 (3) (2017) 032006.
[10] I. Coarasa et al., ANAIS-112 sensitivity in the search for dark matter annual modulation, Eur. Phys. J. C. 79 (2019) 233.
[11] M. Antontello et al., The SABRE project and the SABRE Proof-of-Principle, Eur. Phys. J. C 79 (2019) 363.
[12] K. Fushimi, et al., Dark matter search project, PICO-LON, J. Phys. Conf. Ser. 718 (4) (2016) 042022. doi:10.1088/1742-6596/718/4/042022
[13] G. Angloher et al., The COSINUS project: perspectives of a NaI scintillating calorimeter for dark matter search, Eur. Phys. J. C 76 (2016) 441.
[14] R. Bernabei, et al., Final model independent result of DAMA/LIBRA-phase1, Eur. Phys. J. C 73 (2013) 2648. doi:10.1140/epjc/s10052-013-2648-7
[15] R. Bernabei, et al., First model independent results from DAMA/LIBRA-Phase2, Nucl. Phys. At. Energy 19 (2018) 307, arXiv:1805.10386 [astro-ph.IM].
[16] S. Agostinelli, GEANT4: a simulation toolkit, Nucl. Instrum. Meth. A 506 (2003) 250, doi:10.1016/S0168-9002(03)01368-8. doi:10.1140/epjc/s10052-018-5970-2
[17] P. Adhikari et al. (COSINE-100 collaboration), Background model for the NaI(Tl) crystals in COSINE-100, Eur. Phys. J. C 78 (2018) 490. doi:10.1140/epjc/s10052-018-5970-2
[19] E. Barbosa de Souza, et. al. (COSINE-100 collaboration), Study of cosmogenic radionuclides in the COSINE-100 NaI(Tl) detectors, Astropart. Phys. 115 (2020) 102390.

[20] J. Amare, et. al., Analysis of backgrounds for the ANAIS-112 dark matter experiment, Eur. Phys. J. C 79 (2019) 412.

[21] J. Amare, et. al., Assessment of backgrounds of the ANAIS experiment for dark matter direct detection, Eur. Phys. J. C76 (2016) 429.

[22] S. Cebrian, et. al., Background model for a NaI (Tl) detector devoted to dark matter searches, Astropart. Phys. 37 (2012) 60.

[23] N. J. T. Smith, J. D. Lewin, P. F. Smith, A possible mechanism for anomalous pulses observed in sodium iodide crystals, Phys. Lett. B 485 (2000) 9.

[24] S. Cooper, H. Kraus, J. Marchese, Radon-implanted $^{214}$Po and anomalous pulses in sodium iodide detectors for dark matter, Phys. Lett. B 490 (2000) 6.

[25] R. Agnese, et. al., Demonstration of surface electron rejection with interleaved germanium detectors for dark matter searches, Appl. Phys. Lett. 103, 164105 (2013).

[26] R. Agnese, et. al., Projected sensitivity of the SuperCDMS SNOLAB experiment, Phys. Rev. D 95 (2017) 082002.

[27] R. Strauss, et. al., A detector module with highly efficient surface-alpha event rejection operated in CRESST-II Phase 2, Eur. Phys. J. C 75 (2015) 352.

[28] G. Adhikari, et al., Initial performance of the COSINE-100 Experiment, Eur. Phys. J. C 78 (2018) 107. doi:10.1140/epjc/s10052-018-5590-x

[29] G. Adhikari, et al., An experiment to search for dark-matter interactions using sodium iodide detectors, Nature 564 (7734) (2018) 83–86. doi:10.1038/s41586-018-0739-1

[30] S. C. Kim et al., New Limits on Interactions between Weakly Interacting Massive Particles and Nucleons Obtained with CsI(Tl) Crystal Detectors, Phys. Rev. Lett. 108 (2012) 181301, 1204.2646.

[31] K. W. Kim et al., Measurement of low-energy events due to $^{222}$Rn daughter contamination on the surface of a NaI(Tl) crystal, Astropart. Phys. 02 (2018) 51, 1801.06948.

[32] Pin Yang, Charles D. Harmon, F. Patrick Doty, and James A. Ohlhausen, Effect of humidity on scintillation performance in Na and Tl activated CsI crystals, IEEE Trans. Nucl. Sci. 61 (2) 2014.