Incidental radioactive background in BGO crystals

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In this paper some cases of unusual internal radioactive background in BGO crystals are described. Routinely produced at the Nikolaev Institute of Inorganic Chemistry during nearly quarter of century BGO crystals have low radioactive background, caused by ^{207}Bi contamination. But a few batches of BGO crystals incidentally have higher internal radioactive background with activity up to 10 Bk/kg related with other contaminations. One type of the background is pure alpha radioactivity. It is caused by ^{210}Po contamination and has technogenic origin. The other background is identified as gammas coming from short living ^{214}Bi and ^{214}Pb isotopes. It indicates the pollution of crystals by long living isotopes, most likely ^{226}Ra. Unusually high rate suggests a high probability of technogenic origin of this background too.

1. Introduction

BGO ($Bi_4Ge_3O_{12}$) is one of the most commonly used scintillating crystals. Its components have not long living radioactive isotopes. Recently discovered instability of ^{209}Bi itself causes a negligible background rate because of very long decay time ($T_{1/2}=2\times10^{19}$ years) [1]. The main source of the internal radioactive background of the BGO crystals is contamination by ^{207}Bi with typical activity 1–3 Bk/kg [1, 2, 3, 4]. It corresponds to ^{207}Bi concentration of order of 10^{-15} g/g. A study of the dependence of ^{207}Bi contamination on the source of bismuth shows that using bismuth from lead ore mines creates an order of magnitude higher gamma background compared to lead free mines [2]. Therefore the author suggests the hypothesis of ^{207}Bi production via interaction of the cosmic rays protons with lead $^{206}Pb+p\rightarrow^{207}Bi$. A measurement of ^{207}Bi background on earth surface and in deep salt mine shows the same rate [3]. It eliminates the hypothesis of gamma pollution from direct activation of ^{209}Bi in a crystal by cosmic muons via reaction $^{209}Bi(\mu^-,2n)^{207m}Pb$ ($T_{1/2}=0.8$ s). The upper limits on the contamination by ^{238}U and ^{232}Th are set as low as 10^{-11} g/g [3]. It corresponds to 2–3 orders of magnitude lower radioactive background compared to ^{207}Bi activity.

BGO crystals are grown at the Nikolaev Institute of Inorganic Chemistry (NIIC) from beginning of 1980-s [5, 6]. Contamination by ^{207}Bi is usually low. However some crystal batches have a substantial intrinsic radioactive background from other sources. In this paper, the types of backgrounds have been identified as gamma emitting isotopes for some crystals and pure alpha radioactivity for others.

2. Gamma radioactive background

The main source of gammas in ^{207}Bi decay is cascade transition $^{207m}Pb\rightarrow{}^{207}Pb$. It gives single peaks at 570, 1063 keV and sum peak at 1633 keV in BGO background spectrum.

The high background rates about 10 Hz/kg with lines similar to ^{207}Bi were observed in one batch of the BGO crystals in 1999. The gamma energies from ^{214}Bi decays (609, 1120 and 1764 keV) are close to ones from ^{207}Bi (Fig. 1). The lower energy lines from ^{214}Pb decays in gamma

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spectra are hidden under Compton tails and other low energy background. So it is difficult to discriminate those isotopes from intrinsic low rate background spectra of BGO crystals. The gamma spectrum from intrinsic background radiation of the BGO crystal has been measured with the low background HPGe detector. The lines 350 keV from ^{214}Pb and 609 keV from ^{214}Bi are clearly seen. No associated with ^{207}Bi gamma lines have been observed in the spectrum.

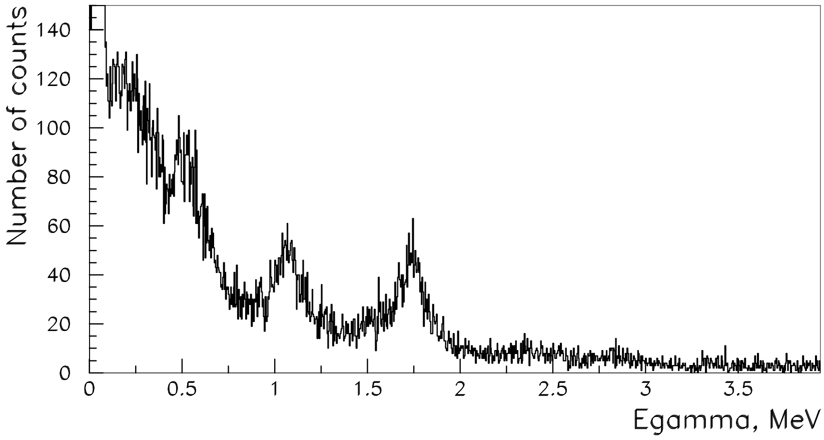

Fig. 1 Sample of intrinsic gamma background in BGO crystal.

Both ^{214}Pb and ^{214}Bi isotopes are far from stable isotopes and have short decay time ($T_{1/2}$= 27 and 20 minutes respectively) to be produced by cosmic ray interactions. Besides they are part of the natural radioactive background coming from ^{238}U decay chain. Therefore a presence of ^{214}Pb and ^{214}Bi isotopes indicates a pollution of the crystals by long living isotopes from ^{238}U decay chain. To explain observed gamma background rate the ^{238}U concentration should be of order of 10^{-6}–10^{-7} g/g. It is unlikely to be true. Such level of contamination will affect optical properties of a crystal. More reasonable hypothesis is ^{226}Ra contamination at level of a few units of 10^{-13} g/g. The alphas from radium decay chain give energy deposition in BGO equivalent to slightly more than 1 MeV gammas because of quenching factor of about 5. Therefore alpha lines merge with the 1120 keV gamma line.

We suggest the following hypothesis. During raw material purification and crystallization processes bismuth is cleaned from uranium and thorium but not from radium. An indirect support of this hypothesis comes from BGO crystal alpha background spectrum in Ref. [1]. There are no ^{238}U and ^{235}U lines and only trace amount of ^{232}Th in this spectrum. But alpha lines from radium isotopes and their decay products are clearly seen. Anyway the pollution by natural radioactive isotopes in this crystal is orders of magnitude higher than usually. A source of pollution is untraceable but its large level suggests high probability of its technogenic origin.

3. Pure alpha radioactive background

A background counting rate of about 10 Hz/kg was observed in some crystals. The spectrum shows a clear peak with energy deposition equivalent to about 1 MeV gammas (Fig. 2). This peak has no associated Compton part of spectrum. It indicates not a gamma origin of the peak but alpha.

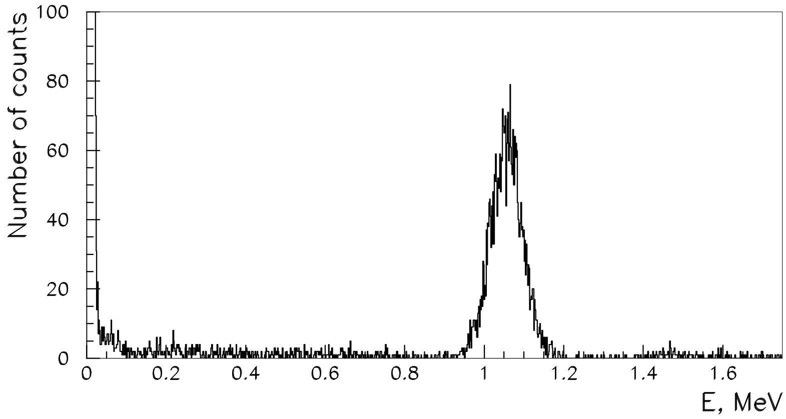

Fig. 2. Intrinsic BGO background spectrum.

To check the alpha hypothesis a background free crystal has been coupled to a PMT and the suspected crystal has been placed on top of it as radioactive source (Fig. 3). No additional rate was observed. Measurement with a low background HPGe detector shows no additional gamma background too. It demonstrates that radiation particles do not leave the crystal. That supports the alpha origin of the background.

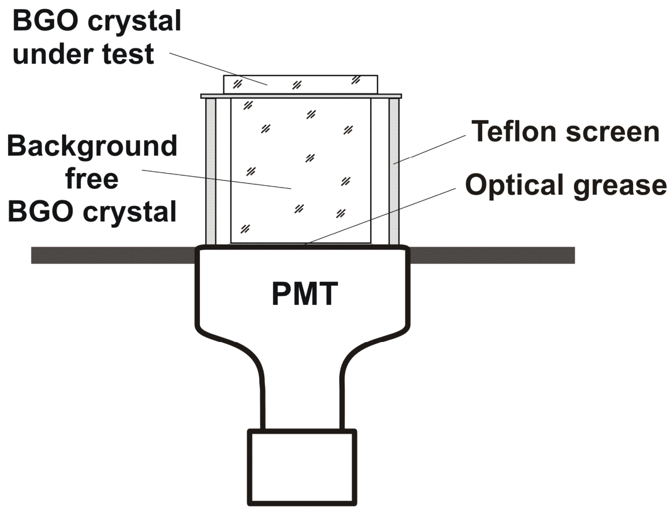

Fig. 3. Scheme of a background set-up with 2 BGO crystals.

The typical energy of alpha particles is several MeV. But response to alpha particles in heavy scintillating crystals is much lower compared to gammas because of quenching effect. This effect is nonlinear. So the peak position cannot be used to identify source of alpha particles by measurement of an energy deposition in BGO crystal. On another hand the absence of gamma rays requires that radioactive series must be short, preferably consisting of one nuclide. The good candidate is ^{210}Po isotope. The measured with alpha spectrometer energy of emitted alphas is 5.3 MeV in accordance with ^{210}Po radiation. The observed background rate corresponds to ^{210}Po pollution lower than 10^{-16} g/g.

The absence of gamma rays shows rather neutron activation origin of ^{210}Po isotope due to reaction chain ^{209}Bi+n → ^{210}Bi (β$^-$, 5 days) → ^{210}Po than natural radioactivity. The ^{214}Bi and ^{214}Pb gamma lines must be present in case of uranium or radium origin of the polonium. Pure Bi metal for Bi oxide production is refined at NIIC from commercial Bi, having 99.9% purity. Residuals from electrolyzes (one stage of refining) are enriched by possible radioactive impurities. The gamma spectrum of selected residuals was measured with Ge-Li detector. Many associated with ^{152}Eu lines are seen (Fig. 4). This isotope has half life of 13 years and is absent in natural isotope composition.

But ^{151}Eu has large cross section of neutron activation (σ=9200 b). So the tiny amount of ^{151}Eu can produce substantial radioactive background after neutron irradiation. The presence of the radioactive europium isotope confirms hypothesis of technogenic origin of the ^{210}Po contamination.

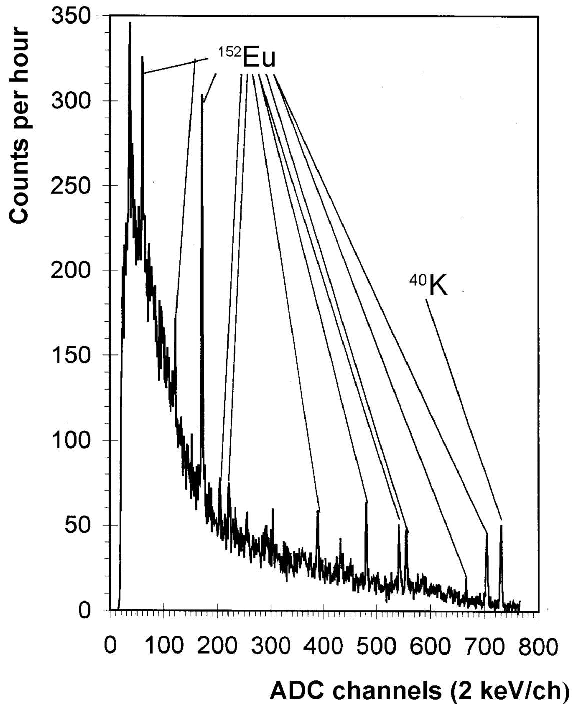

Fig. 4. Gamma spectrum of Bi electrolyzes residuals measured with Ge-Li detector.

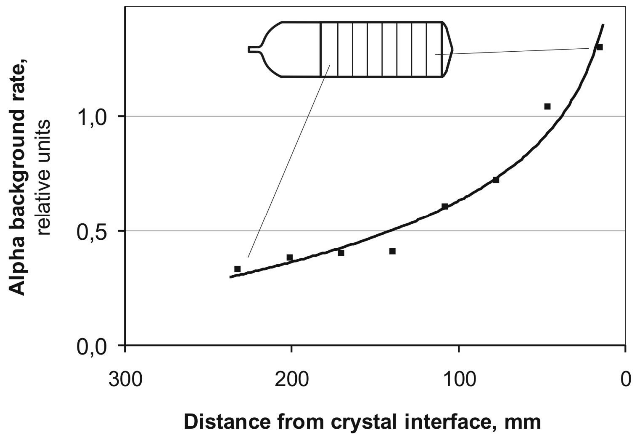

Fig. 5. Alpha background rate vs distance from crystal interface.

The measurements of alpha background rate along the crystal show an increasing rate from beginning to the end of crystal (Fig. 5). It means enhancement of the ^{210}Po concentration in melt and decreasing in grown crystals. Therefore BGO crystals could be purified from ^{210}Po contamination by recrystallization. To check this, high alpha background BGO crystals were selected and used as charge to grow a new BGO crystal. The background activity of re-grown crystal is about 3.5 times less than that of initial crystals. So recrystallization of BGO crystals decreases alpha background.

So far we know there is no literature data on this type of BGO radioactivity. First time it was seen in 2003. However, it happens more and more frequently later and can become a problem for low background experiments. A probable reason is following. The main source of bismuth is lead production industry. Production of lead decreases now because of ecology reasons [7]. Therefore more and more bismuth on market comes from recycling and not from primary production. The bismuth is widely used at nuclear power stations. Observed radioactivity is small from point of view of safety rules. So polluted by ^{210}Po bismuth can come from utilization of used Bi at nuclear power stations.

4. Conclusion

The BGO crystals grown in the Nikolaev Institute of Inorganic Chemistry have a low intrinsic radiation background caused by ^{207}Bi contamination. However a small fraction of the crystals have a substantially higher background counting rate. Two types of background were found. The first one is a pure alpha background caused by ^{210}Po isotope contamination. This contamination comes most probably from neutron irradiation. So it has technogenic origin. The second one is gamma background coming from short living ^{214}Pb and ^{214}Bi isotopes. They are part of natural radioactive background and indicate the presence of long living isotopes, most likely ^{226}Ra. Moreover, the unusually large concentration of those isotopes suggests a high probability of technogenic origin. The precise sources of radioactive contamination are untraceable. One possible explanation is the recycling of bismuth used at nuclear power stations. Primary bismuth from lead free ore mines should be used to grow BGO crystals for low background applications.

References

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