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Limits and performances of a $BaWO_4$ single crystal

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ABSTRACT

A Barium Tungstate single crystal ($BaWO_4$) was produced using the low thermal gradient Czochralski technique. In this paper the results concerning its light emission and radioactive contaminants are presented. The aim of this work is to investigate the possibility to realize $BaWO_4$ crystals with suitably features to study the double beta decay of ^{130}Ba and ^{132}Ba isotopes by the "source = detector" approach. The results show the limitations of a $BaWO_4$ crystal as a scintillator and give some idea on how to overcome them in order to take profit from the potentiality of this single crystal.

1. Introduction

The double beta decay with emission of two neutrinos ($2\nu2\beta$) [1] is a nuclear transition allowed in the Standard Model (SM) of particle physics and observable in many even–even nuclei. This transition has so far been measured for several nuclei [2]. Moreover, neutrinoless double beta ($0\nu2\beta$) decay [3] is forbidden in the theoretical framework of the SM; it is, however, predicted in several theoretical SM extensions [4–7]. Therefore, $0\nu2\beta$ decay is a very suitable approach to investigate some of the major unresolved issues in particle physics, such as lepton number conservation, Majorana nature of neutrinos, and neutrino masses. In addition, experimental data on the $2\nu2\beta$ decay could be very useful to improve theoretical calculations of the decay probability related e.g. to the nuclear matrix elements [5].

Natural barium contains two potentially 2β active isotopes,^{130}Ba ($Q_{2\beta}$ = 2618.7(2.6) keV) and ^{132}Ba ($Q_{2\beta}$ = 844.0(1.1) keV) [8,9]. The ^{130}Ba isotope is of particular interest because its double beta decay is not yet detected unambiguously; at present there is only an indication of a multichannel weak decay ($2\beta^+$, $\epsilon\beta^+$ and 2ϵ) in ^{130}Ba by geochemical experiments with determined values of the half-life of $T_{1/2} = (2.2 \pm 0.5) \times 10^{21}$ y [10] and $T_{1/2} = (6.0 \pm 1.1) \times 10^{20}$ y [11]. The first direct laboratory search for 2β decays of ^{130}Ba was performed by using a BaF_2 crystal scintillator [12], where only $T_{1/2}$ limits were obtained at the level of $\approx 10^{17}$ yr. In this frame, the development of a Ba-containing crystal scintillator is a very powerful tool to investigate through the low-background "source = detector" approach the two-neutrino and zero-neutrino double-beta decay in Ba isotopes.

There are more than 60 Ba-containing compounds that have been investigated as crystal scintillator by different groups. A detailed list of these compounds and their main operational properties is reported in [13]. In Table 1, instead, we have listed just the most prospective ones from the point of view of light yield and radiopurity. In this paper we report on recent progress in the production of high quality, large volume $BaWO_4$ single crystals using the Low Thermal Gradient (LTG) Czochralski method with highly purified material—WO_4 powder [14–20]. The combination of these factors looks very promising for the production of high quality $BaWO_4$ given the long history of success producing WO_4-based crystal [19,21–24,14].

2. Development of a commercial $BaWO_4$ crystal

One possible way to produce low-background high quality crystals is to use the LTG Czochralski technique [55–57], which operates with a closed Pt-crucible. An additional and very important advantage of this

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Table 1
Inorganic Ba-based scintillating compounds and some of their optical properties [13].

Formula(*dop.*)	Light yield (photons/keV)	λ Emission peak (nm)	Reference
$Ba_2GdCl_7(Ce)$	30	355; 377	[25]
$Ba_2Si_3O_8(Eu)$	35	505	[25]
$Ba_2SiO_4(Eu)$	22; 40	502; 505; 525	[25,26]
$Ba_3(PO_4)_2(Eu)$	27	42	[25]
$Ba_3P_4O_{13}(Eu)$	25	440	[25]
$Ba_5Si_8O_{21}(Eu)$	20	511	[25]
$BaBr_{1.7}I_{0.3}$	112	414	[27]
$BaBr_2$	19.3	425; 475	[28]
$BaBr_2(Ce)$	~10.3	345; 370	[29,30]
BaBrCl(*Eu*)	~45	405	[31,32]
BaBrI(*Eu*)	85	413	[33,34,27,35]
$BaCl_2(Eu)$	19; 52	~400	[28,30,36,37]
BaClBr(*Eu*)	52		[38]
BaClI(*Eu*)	54		[38]
BaF_2	1.3–63.9	~200; 310; ~360	[39–46]
BaFI(*Eu*)	55	405	[47]
$BaGdCl_5(Ce)$	35	363; 389	[25]
$BaHfO_3(Ce)$	40	400	[48]
$BaKPO_4(Eu)$	35	425	[25]
$BaSi_2O_5(Eu)$	30	520	[25]
$Cs_2BaBr_4(Eu)$	~25		[38]
$Cs_2BaCl_4(Eu)$	~30		[38]
$Cs_2BaI_4(Eu)$	~17		[38]
$CsBa_2Br_5(Eu)$	~70		[38,49]
$CsBa_2I_5(Eu)$	~90	435	[50,34,35,51]
$CsBa_2I_5(In)$	35	540	[52]
$CsBa_2I_5(Na)$	33	430	[52]
$CsBa_2I_5(Tl)$	40		[52]
$CsBa_2I_5(Yb)$	54		[53]
$K_2BaI_4(Eu)$	63	448	[54]
$KBa_2I_5(Eu)$	90	444	[54]
$NaBaPO_4(Eu)$	20	450; 610	[25]

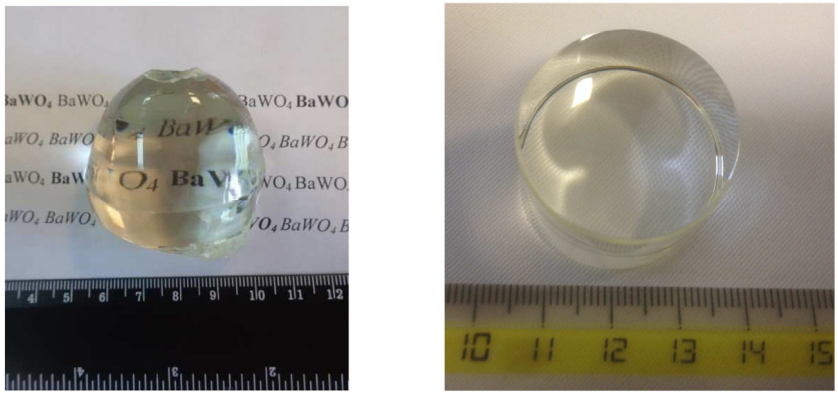

Fig. 1. (Left) $BaWO_4$ boule (about ø45 mm × 45 mm). (Right) $BaWO_4$ single crystal used in the present paper (ø28 mm × 23 mm). No inclusions, visible defects and cracks into the crystal volume have been observed.

technique is the small temperature gradient at the level of about 1 K/cm, which is a one to two orders of magnitude lower than the conventional Czochralski method where it can reach 60–100 K/cm. Due to the low temperature gradient, and therefore low evaporation of material from the melt, the losses do not exceed 0.5% of the initial charge. Last, but not the least, the LTG Czochralski technique allows to crystallize up to 90% of the loaded charge (already achieved for BGO [58], cadmium tungstate [14,59] and zinc molybdate [60] crystals). This feature is crucial in case of costly materials, containing enriched isotopes or highly purified initial components. The $BaWO_4$ single crystal we used in this study was grown using the LTG Czochralski technique at the V.S. Sobolev Institute of Geology and Mineralogy of the Siberian Branch of the Russian Academy of Sciences (Novosibirsk, Russia). To produce the $BaWO_4$ crystal, a furnace with resistive heating and a configuration similar to the one described in [61] were used. This configuration with a three-zone heater allows to change the thermal conditions within wide limits and to ensure an almost flat front of crystallization. The best results were obtained with a slightly convex crystallization front and a crystallization rate of 1–1.5 mm/h.

The tungsten oxide powder (WO_3), which was additionally purified similar to that used for the production of $ZnWO_4$ [62] and $^{116}CdWO_4$ [63] crystals, and commercially available barium carbonate ($BaCO_3$) of 99.5% purity grade were used as initial components for the crystal production charge. The stoichiometric mixture of the initial components was loaded into a platinum crucible ø70 mm × 150 mm, and gradually heated with a rate of 100 K/h up to the melting point (about of 1500 °C) [64]. The total duration of the homogenization and synthesis processes was about 10 h. Despite the relatively high temperatures applied, the platinum crucible remained in the air atmosphere without significant changes in geometry and weight. The total mass of loaded $BaWO_4$ charge in the crucible was 550 g, obtaining eventually a $BaWO_4$ single crystalline boule – without visible