



Multi-ampoule Bridgman growth of halide scintillator crystals using the self-seeding method



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ABSTRACT

We investigate the multi-ampoule growth at 25 mm diameter of ternary iodide single crystal scintillator $\text{KCaI}_3\text{:Eu}$ using the randomly oriented self-seeded Bridgman method. We compare scintillation performance between cubic inch scale crystals containing small variations of low nominal europium concentrations previously shown to balance light yield with self-absorption in the host crystal. Growth conditions were optimized in the developmental furnace and four 2 in³ $\text{KCaI}_3\text{:Eu}$ crystals were grown simultaneously producing a total of six 25 mm × 25 mm cylinders. Small variations in activator concentration did not result in significant performance differences among the six measured crystals. A range of energy resolutions of 3.5–4.7% at 662 keV was achieved, surpassing that of NaI:Tl crystals commonly used in spectroscopic detection applications. The function and basic design of the multi-ampoule furnace as well as the process of growing single crystals of KCaI_3 is included here.

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1. Introduction

Halide based single crystal scintillators are widely used in spectroscopic detectors of ionizing radiation such as X-rays and gamma-rays in order to identify and intercept radiological materials. Materials used within the detectors in these applications must have excellent energy resolution in order to distinguish the signatures of special radiological material and naturally occurring radiation (background). Halide scintillators such as thallium doped NaI (NaI:Tl) and CsI (CsI:Tl) can attain energy resolutions of 6–7% at 662 keV. These materials have been in use for decades and their growth processes using variations of the Bridgman or Czochralski methods have been well developed, making the costs low enough for widespread use. Recently an emphasis has been placed upon enhancing isotope identification technology as a response to the increasing threat of nuclear attacks on a global scale and in support of enforcing nuclear non-proliferation treaties. This effort is reliant upon development of cost effective alternatives to the superior spectroscopic capabilities of semiconductor technologies such as high purity germanium (HPGe) based detectors [1]. Room-temperature operating direct-collection semiconductor detectors such as cadmium zinc telluride (CZT) have also been in development for some time [2]. However, despite energy resolution of ≈1% or better at 662 keV, the combination of special operating con-

ditions, the extremely high purity required of these materials, and the difficulties encountered in crystal growth results in high costs and limited availability for widespread deployment.

Development of high performance scintillators such as $\text{LaBr}_3\text{:Ce}$ [3] and $\text{SrI}_2\text{:Eu}$ [4] has resulted in energy resolutions in the 3% range thereby bridging the performance gap between existing semiconductor based detectors and scintillator based detectors using NaI:Tl and CsI:Tl . Nonetheless, these new materials suffer from high costs that can be attributed in part to lower yields of single crystal growth processes and the high cost of raw materials. Consequently, present efforts on behalf of a great many research entities spanning national labs, private industry, and research universities are engaged in the search for yet another generation of scintillator and semiconductor materials with a greater emphasis on cost reductions to improve the commercial availability.

Challenges to scintillator production can be generally divided into those encountered in the growth of large (1 in³ scale) single crystal monoliths, and those pertaining to achieving uniform composition/performance characteristics. In the case of scintillation materials used in a spectroscopic detector, the crystals must be of sufficient optical quality and free of significant inclusions and cracking to ensure efficient and uniform light collection. The achievable energy resolution is the most important metric to isotope identification which is primarily a function of the scintillation light yield, non-proportionality of response to a range of excitation energies, and uniformity of scintillation light production and collection within the crystal and photosensor respectively [5]. For

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activated or extrinsic scintillators, the uniformity of scintillation light production relies upon the homogeneity of defects and activator species throughout the crystal bulk.

Until recently, Potassium containing scintillators have been overlooked for their use in radioisotope identification devices due to the drawbacks associated with the natural abundance of ^{40}K . Investigations into the ternary iodide materials such as $\text{KCaI}_3:\text{Eu}$, $\text{K}(\text{Ca,Sr})\text{I}_3:\text{Eu}$ and $\text{KSrI}_5:\text{Eu}$ have shown these compounds to share desirable properties with high scintillation light yields and capable of attaining energy resolution in small crystals of $\approx 3\%$ or better at 662 keV [6–8]. Scale-up efforts have shown cubic inch scale crystals of these materials maintain the excellent light yield and energy resolution and warrant their continued development [9–11]. The presence of ^{40}K in these materials produces a pronounced beta background along with the characteristic 1460 keV gamma photopeak in pulse-height spectra and negatively affects time-to-detection figure of merit for weak source activity. In $\text{LaBr}_3:\text{Ce}$ and CeBr_3 scintillators, a similar elevated background due to natural abundance of ^{138}La and actinium impurities hinder their use in low count rate applications but are desirable in applications requiring excellent energy resolution in high source activity environments [12]. Sourcing material with reduced concentrations of the ^{138}La isotope and improved processing have been effective in reducing the elevated background in these materials [13].

Development of Bridgman growth processes at the cubic inch scale for $\text{KCaI}_3:\text{Eu}$ has made rapid progress resulting from extensive growth experiments in transparent and conventional furnaces. As a result, this composition has been the focus of current efforts to investigate its potential for further development. One challenge encountered for scale-up of europium doped scintillators is maintaining performance as the crystal volume is increased due to complications arising from self-absorption [14]. A previous examination into the effects of increasing crystal size and europium concentrations in this compound show self-absorption can be minimized with use of low Eu doping in the 0.5–1.0 at% range. Use of lower Eu dopant can maintain shorter scintillation lifetimes on the order of 2 μs and maintain excellent energy resolution in the 4% range for cubic inch scale crystals.

Optimized growth conditions for KCaI_3 single crystals have been incorporated within a multi-ampoule growth station (MAGS) developed at our research facility at the University of Tennessee. The developmental Bridgman-type furnace adopts the concept of growing multiple boules of smaller diameter simultaneously when it is economically unfeasible, or exceedingly difficult, to produce the same volume and quality of crystal in one boule with a larger diameter. Such multi-ampoule furnace designs incorporating an inductively or resistively heated crucible for growth of GaAs as well as fluoride based materials have been proposed [15–17]. Similarly, multi-ampoule furnaces have been designed at the Shanghai Institute for Ceramics, Chinese Academy of Sciences (SICCAS) for large-scale production of $34 \times 34 \times 360$ mm PbWO_4 crystals for use as detectors in electromagnetic calorimeters [18], as well as piezo electric crystals $\text{Li}_2\text{B}_4\text{O}_7$ (at $\varnothing 105$ mm) [19] and $\text{Sr}_3\text{Ga}_3\text{Ge}_4\text{O}_{14}$ (at $\varnothing 50$ mm) [20]. In each case of the SICCAS developed furnaces, the conventionally cylindrical bore is instead elongated to accommodate multiple ampoules in a side by side configuration, all using a single crystal seed fitted in the bottom of the ampoule to promote growth of a single grain of a desired orientation. This approach was successful in each case in producing multiple single crystal boules of high quality.

To our knowledge, this modification of the vertical Bridgman process has not yet been employed in the growth of iodide based crystals. One major difficulty in adapting the process to iodide crystal growth is the fabrication and handling of seed crystals with a desired orientation from the typically hygroscopic and brittle single crystals. During the course of our research surrounding KCaI_3

utilizing the MAGS we have exclusively employed growth using a randomly oriented self-seeded approach using grain isolating capillary geometries [21,22].

In this work we compare scintillation performance between cubic inch scale crystals of $\text{KCaI}_3:\text{Eu}$ grown in the MAGS. We examine the effects of varying low nominal europium concentrations on the basis of spectroscopic performance, namely energy resolution, as well as scintillation decay time. We will illustrate the basic design and functionality of the MAGS and procedures used for growth of multiple single crystal boules of $\text{KCaI}_3:\text{Eu}$ at $\varnothing 25$ mm, simultaneously, using a randomly oriented self-seeded approach. Obstacles encountered in optimizing this process for $\text{KCaI}_3:\text{Eu}$ will also be discussed.

2. Equipment and experimental methods

2.1. The multi-ampoule growth station

The multi-ampoule growth station (MAGS) was designed and constructed using widely available ceramic materials and industrial grade power handling components. The furnace has a maximum operating temperature of 1000 °C and is divided into two independently controlled heating zones separated by a 2.5" insulated region housing a diaphragm. The basic construction is comprised primarily of firebrick supported by a steel frame. Use of the diaphragm between the heating zones is critical in maintaining a stable thermal gradient due to the large size of the furnace bore [23]. Heating is achieved by 12 easily replaceable silicon carbide bar elements with an effective heating region of 18" to match the rectangular bore. The crystal lowering mechanism, and each heating zone is programmed and operated using a logic controller and graphical user interface.

The translation stage is driven by a heavy duty stepper motor based system and houses hardware for supporting an accompanying ampoule vessel refractory tube for each ampoule bay. The stage and diaphragm can be reconfigured for use of fewer or more ampoule bays. Each ampoule vessel support post can be independently manipulated for small adjustments to the vertical position of each ampoule vessel and for loading and unloading the furnace. Each ampoule vessel can fit up to a $\varnothing 2$ " diameter ampoule which is supported by a pourable insulation comprised of fine alumina grog of 1–2 mm in size. To aid in monitoring the seeding and growth processes, thermocouples embedded into each ampoule vessel indicate temperatures at the seed and last-to-freeze regions of the charge. The signal from the embedded thermocouples can be used to trigger the advancement of a growth program in order to automate the entire process once the starting charges are loaded into the furnace. An illustration of the MAGS is shown in Fig. 1.

2.2. Experimental procedure for multi-ampoule growth of $\text{KCaI}_3:\text{Eu}$

A polycrystalline ingot is prepared in a round bottom fused silica ampoule by loading with a stoichiometric ratio of $\text{Kl}:\text{CaI}_2(1-x):\text{EuI}_2(x)$ with x ranging from 0.005 to 0.01. This level of doping has been observed to produce an optimal balance between scintillation light yield and self-absorption effects [8]. The precursor materials purchased from APL Engineered Materials Inc. are anhydrous metal halide and rare-earth halides with 99.995–99.999% purity (trace metals basis) and in beaded form (10 mesh). All handling of the halide materials before and after synthesis and growth is performed in a dry nitrogen glovebox maintained below 0.1 ppm oxygen and moisture to prevent decomposition and/or moisture absorption.

Once loaded, the ampoule is transferred to a vacuum furnace where the precursors are dried for several hours above 200 °C

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