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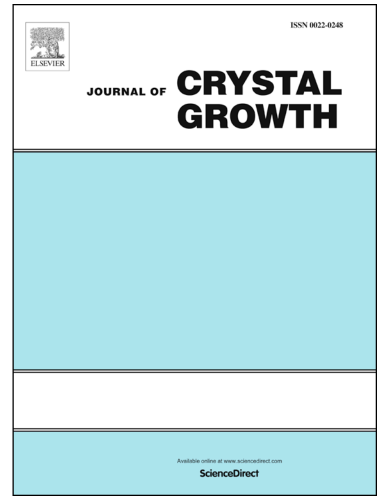

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ACCEPTED MANUSCRIPT

Advanced Crystal Growth Techniques for Thallium Bromide Semiconductor Radiation Detectors

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ABSTRACT

Thallium Bromide (TlBr) is a promising room-temperature radiation detector candidate with excellent charge transport properties. Currently, Travelling Molten Zone (TMZ) technique is widely used for growth of semiconductor-grade TlBr crystals. However, there are several challenges associated with this type of crystal growth process including lower yield, high thermal stress, and low crystal uniformity. To overcome these shortcomings of the current technique, several different crystal growth techniques have been implemented. These include: Vertical Bridgman (VB), Physical Vapor Transport (PVT), Edge-defined Film-fed Growth (EFG), and Czochralski Growth (Cz). Techniques based on melt pulling (EFG and Cz) were demonstrated for the first time for semiconductor grade TlBr material. The viability of each process along with the associated challenges for TlBr growth has been discussed. The purity of the TlBr crystals along with its crystalline and electronic properties were analyzed and correlated with the growth techniques.

Keywords: A2. Bridgman technique; A2. Edge defined film fed growth; A2. Czochralski method; A2. Growth from vapor; B3. Radiation detectors; B1. Thallium Halides

1. INTRODUCTION

Thallium Bromide (TlBr) is a promising room-temperature wide bandgap semiconductor radiation detector material. Compared to other semiconductors, TlBr has higher photoelectric and total attenuation coefficients for gamma rays with energy greater than 20 keV [1]. The cubic crystal structure and fairly low growth temperature of this binary compound (480 °C) significantly simplifies the crystal growth process. Furthermore, TlBr is self-compensated and shows high electrical resistivity (10^{10} – 10^{11} Ωcm) without doping. These give TlBr a distinctive advantage over other high melting temperature ternary semiconductors (such as the flagship candidate Cadmium Zinc Telluride).

The primary technique for purification and growth of TlBr is the Travelling Molten Zone (TMZ) technique [2-11]. The zone refining purification technique improves the charge transport properties [4,7], and hence is an integral part of the entire growth process. The TMZ technique, however, suffers from a number of drawbacks. First, as the grown crystal boule is in the shape of a semi-cylinder, a non-trivial portion of the boule volume cannot be used for device fabrication. It leads to lower effective yield and higher per cc cost of the detectors. Second, there is a significant thermal stress on the growing TlBr crystal [11]. This, in turn, affects the properties of the detectors fabricated from these crystals. Using TMZ, we have grown single crystalline TlBr crystals up to 2-inches in diameter. Due to the asymmetry in the radial temperature gradient, this is the largest size of the TlBr crystals that can be grown using this technique. With a larger diameter, it is impossible to maintain the crystallinity and quality of the growing crystal.

In this paper, we report on the crystal growth of TlBr by the Vertical Bridgman (VB), Physical Vapor Transport (PVT), Edge-defined Film-fed Growth (EFG) and Czochralski Growth (Cz) techniques and compare the properties of the grown crystals with those grown by the standard TMZ process.

2. EXPERIMENTS

The TMZ crystals are generally grown in two configurations, namely closed and open ampoules. In the closed ampoule configuration, the high purity (5N) TlBr beads are sealed inside a quartz ampoule with a reactive atmosphere, typically consisting of HBr, H_2, and an inert gas. The charge is horizontally zone refined multiple times (usually ~50-100) at a rate of 5cm/hr. HBr is used as a reducing agent for removal of oxygen and hydroxyl-ions, and to maintain the stoichiometry of the TlBr melt [7]. The charge translation rate is lowered to 0.2cm/hr in the last run to achieve single crystal growth of the boule. In the open ampoule configuration, the same steps as in the close ampoule approach are followed, except that the reactive gas mixture is continuously flown over the charge. The flowing gas removes any impurities that may be rejected from the molten zone in the ampoule. However, because molten TlBr has a relatively high vapor pressure, some of the TlBr charge is transported out of the ampoule reducing the overall process yield. In both the closed and open ampoule TMZ runs, the two ends of the charge (1.5-inch each) which have a high impurity concentration were discarded and detectors were fabricated from the central region of the boule.

For the VB growth, the charge was initially zone refined in a closed ampoule and the central portion of the purified charge was transferred to a second quartz ampoule with a conical bottom. The ampoule was placed in a two-zone furnace, the charge was melted and superheated to 600°C for 24 hours prior to growth. The ampoule was translated at a rate of 10mm/day and solidification occurred under an average temperature gradient of 20°C/inch at the growth interface.

In PVT growth, 5N TlBr beads were first sublimed to remove low vapor pressure impurities. The purified material was then transferred into a two-bulb 1-in diameter ampoule. The ampoule was placed in a vertical two-zone semi-transparent gold furnace. The source was maintained at 422°C and the growing crystal region was maintained at 380°C. Figure 1 (a) shows the experimental set-up for this type of growth.

For EFG growth, 5N TlBr beads were directly used, without purification, to test the viability of this growth. The charge was placed in a 1.5-inch diameter quartz crucible, and capped with a graphite die. A photograph of the growth system is shown

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