CRYSTAL GROWTH AND APPLICATIONS OF MERCURIC IODIDE*

by

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SUMMARY

Mercuric iodide is a high-Z wide bandgap semiconductor ($Eg \simeq 2.14$ eV at RT) which provides, in addition to good electron transport properties, a unique combination of features that make this material the most promising choice as a low noise, room temperature, x-ray and gamma-ray detector.[1,2] Therefore, HgI_2 is quite different from the narrow bandgap elemental semiconductors, e.g., Si and Ge, and also different in its physical and chemical properties from the binary zinc-blende compounds such as GaAs or InP. Mercuric iodide is a highly anisotropic layered material consisting of alternating layers of Hg and I. HgI_2 undergoes a reversible phase transformation at 127°C from the tetragonal red (α-HgI_2) to the orthorombic yellow structure. This phase transformation makes it impossible to grow HgI_2 from the melt despite a relatively low melting point of about 250°C. Although it is possible to grow crystals from solution,[3] vapor-grown crystals exhibit better charge transport properties. The temperature oscillation method (TOM) based on the periodic reversal of the axial temperature gradient between the crystal and the source material is usually employed.[4] However, a more recent trend in the HgI_2 vapor growth technology is to limit the TOM to the nucleation stage only, while growing crystals in a static temperature gradient (Lamonds, in Reference 2). A clear experimental evidence exists that the variation of growth rate results in the morphological instability, especially for large crystals. The reasons for this effect will be discussed in detail.

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DISCLAIMER

The present state-of-art of the synthesis and purification of the starting material is also reviewed. Although the purification methods yielding large detector quality crystals of HgI_2 are well established, the correlation between the nuclear response properties and imperfections, e.g., impurities or native defects, are still a subject of extensive investigations. The most puzzling result is that even the best samples contain tens to many hundreds ppm of impurities, mainly hydrocarbons and elemental Na, K, Aℓ, Ca, Si, and Fe. In order to explain this discrepancy, Nicolau (see Reference 2) suggested that "condensation of impurities in clusters" takes place in HgI_2. Indeed, the very recent spark source mass spectrographic analysis performed by Muheim[5] has revealed segregation of impurities along preferred crystallographic directions. We are presently investigating the nuclear response of samples cut from different sections of the crystal studied by Muheim in order to find a correlation between detector performance and the impurity content. A little more evidence exists on the stoichiometry, i.e., the molar ratio of I_2 and Hg must not deviate from a stoichiometric value of 2 by more than 0.1% for good nuclear performance.[6] However, low temperature photoluminescence studies of Merz, et al (in Reference 2) indicate that a slight excess of iodide is present typically in good HgI_2 detector crystals. Some additional details of these studies allow, however, speculation that the excess iodine species stabilize the crystalline perfection without affecting the electronic properties of the material substantially.

Over the past several years the mechanical properties and dislocation structure of HgI_2 have been studied using a variety of experimental techniques (Milstein, et al, Gits — see Reference 2). The performance of HgI_2 detectors is undoubtedly very sensitive to crystalline imperfections, which can arise during crystal growth and detector fabrication. Specifically, the manual cleavage has been shown to cause severe damage to the crystallographic perfection of HgI_2.[7] Consequently, surface defects inducing charge carrier recombination has been confirmed using refined nuclear measurements.[8] The surface recombination phenomenon is particularly important for the performance of HgI_2 as an x-ray detector, for which the radiation is absorbed at a depth of several microns. Nevertheless, the performance of the HgI_2 gamma-ray spectrometers is gradually approaching that of cryogenically cooled Si(Li) and HPGe detectors (Dabrowski, et al, Reference 2).

Recent success in the development of thick HgI_2 spectrometers (about 1 cm thick and up to 10 cm^3 active volume) for efficient high-energy (up to 1.5 MeV) gamma-ray spectroscopy.[9] Methods of bias, light illumination, and radiation conditioning have been found to improve the energy resolution and the linearity of response of the thick detectors. Possible mechanisms responsible for the conditioning processes are suggested. Additional advances in the use of HgI_2 as portable counters, charged particle and neutron detectors, photodetectors, as well as its applications in x-ray astronomy and fabrication of multielement and position-sensitive arrays are reviewed in detail.

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