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GROWTH OF SINGLE CRYSTALS OF MERCURIC IODIDE (HgI_2) IN SPACELAB III[1,2]

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ABSTRACT

Continued development of a system designed to grow crystals by physical vapor transport in the environment of Spacelab III will be described, with special emphasis on simulation of expected space conditions, adjustment of crystal growth parameters, and on board observation and control of the experiment by crew members and ground personnel.

A critical factor in the use of mercuric iodide for semiconductor detectors of x-rays and gamma-rays is the crystalline quality of the material. The twofold purpose of the Spacelab III experiment is therefore to grow single crystals with superior electronic properties as an indirect result of the greatly reduced gravity field during the growth, and to obtain data which will lead to improved understanding of the vapor transport mechanism

The experiments planned to evaluate the space crystals, including gamma-ray diffractometry and measurements of stoichiometry, lattice dimensions, mechanical strength, luminescense, and detector performance will be discussed.

INTRODUCTION

Single crystal sections of mercuric iodide (HgI_2) can be used as radiation sensitive elements in x-ray and gamma-ray detection systems. Because of the high atomic number of the constituent elements and the large electronic band gap of the material, the detectors are very efficient, can be operated at ambient temperatures and have low power requirements. These properties make mercuric iodide detectors suitable for use in, for example, x-ray fluorescence systems for elemental analysis and medical tomography instrumentation and for the observation of x- and gamma-ray sources in space from space platforms.

The operation of solid state energy dispersive radiation detectors requires that the electronic charges created by the radiation are collected at electrodes on opposite surfaces of the detecting element. This implies, that the detector material should be of the highest possible quality, so that electrically active centers located within the band gap which can trap carriers during the time period of charge collection are minimized. One approach to reach this goal is to purify the crystal growing starting material to the extent possible and to adjust the stoichiometry in order to avoid native defects. In addition, however, it is also desirable to grow a crystal which is free of structural

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defects such as low angle grain boundaries, dislocation networks, localized twin structures and microscopic voids. This implies that the crystal growth process should be as regular as possible.

Single crystals of mercuric iodide are normally grown by physical vapor transport in a closed, evacuated ampoule at approximately 120°C. The crystals have a tetragonal structure, and slip easily in a direction parallel to the c-plane. There are indications that the crystals can plastically deform under the effect of their own weight at the elevated growth temperatures.

The crystal growth experiment at reduced gravity therefore has the purpose of obtaining a crystal with superior structure qualities as a result of a more regular, diffusion controlled vapor transport and the absence of slip dislocation bands.

FURNACE AND GROWTH AMPOULE DEVELOPMENT

To accommodate the Spacelab III facilities, a special furnace has been designed which reflects the basic elements of the usual ground-based furnaces[1]. A sketch of this prototype furnace is shown in Figure 1.

The crystal growing ampoule, located in the center of the furnace, has a cylindrical shape. The bottom surface of the ampoule is indented so that internally it forms a pedestal on which the crystal grows. The indentation fits over a metal support tube, inside of which a Peltier cooler is installed to provide cooling to the growing crystal.

The thermal profile around the ampoule necessary for crystal growth is provided by two independently controlled heating coils. The lower part of the ampoule is heated by means of a circular coil which is covered with a metal heat equalization ring. A helically wound vertical coil provides basically an equally distributed amount of heat along the vertical walls of the ampoule. The combined heat inputs create a temperature profile which has a minimum at approximately half the height of the ampoule. At this level the crystal growth source material accumulates when the center of the pedestal is not cooled by the cold sting. Nucleation and crystal growth can be initiated by a combination of a reduction in the ampoule bottom temperature and activation of the Peltier cooler.

On the top of the helical coil a reflective shield is installed to minimize heat losses. This furnace assembly is covered with a bell jar of high optical quality, so that the growing crystal can be observed through a microscope.

The temperature profile along the walls of the ampoule used to grow the crystals on the ground is created by a combination of radiation and convection effects. During an experiment in the Spacelab, the driving force for convection in the air between the bell jar and the ampoule will be reduced. In addition, the furnace assembly will assume different orientations with respect to the prevalent acceleration vector. In order to evaluate the effects of changed gravity conditions a series of experimental simulations was performed whereby the space between the bell jar and the ampoule was evacuated and the whole furnace assembly was positioned upside down. These experiments showed that the heat supply around the top of the ampoule is not sufficient when there is not a continuous convective supply of hot air from the lower part of the furnace which collects in the upper part of the bell jar and keeps the top of the ampoule hot. This problem was corrected by installation of a lamellar heater on the inside surface of the reflecting dome (not shown in Figure 1). The power input to this heater is slaved to the power controls for the helical heating coil.

(? LAMINAR?)

The normally used ground-based growth ampoules have smooth walls and a flat pedestal. In the initial phase of the crystal growth procedure, the polycrystalline charge is evaporated from the bottom of the ampoule to the minimum temperature zone on the vertical wall, and subsequently a seed is nucleated by

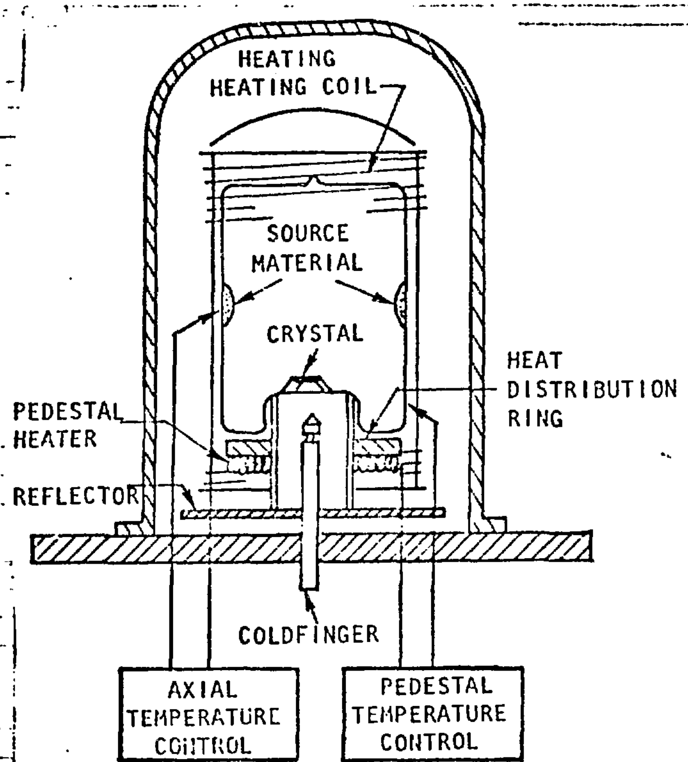

Fig. 1

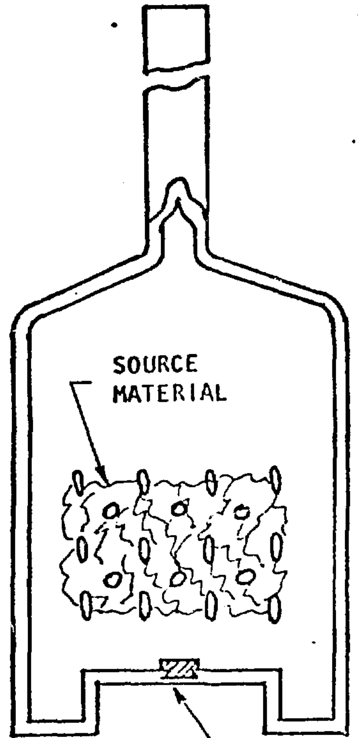

Fig. 2

reducing the pedestal temperature and activating the Peltier cooler. In order to maximize the crystal growth time in space it was decided to perform these activities on the ground before the flight. The resulting ampoules with source and seed in place were subjected to vibration tests, simulating take-off and re-entry vibrations, to ascertain the adherence of source and seed to the glass surface. Both the grown seed and parts of the source material started to come loose at certain critical vibration intensities. The ampoule design was therefore modified to incorporate glass fingers on the part of the inside wall where the source is located and a dovetail cavity in the pedestal to anchor the seed (see Figure 2). The ampoule is positioned in the furnace with the section with glass fingers at the rear. The resistance of the furnace windings has been adjusted in that area so that an asymetric cold region is created for the source material to collect. The seed is nucleated inside the dovetail cavity and is grown until it completely fills and extends somewhat beyond the cavity. The ampoules prepared in this way are again subjected to qualification testing before flight.

CRYSTAL GROWTH PROCEDURE

The crystal growth activities on board Spacelab III will be performed by a crew member who will also be responsible for several other experiments. The procedures to perform the vapor growth experiment have therefore been designed in such a way that a minimal amount of crew activity is required.

During lift-off the furnace assembly is stored in a container which protects it from excessive vibrations. When stable flight conditions are reached, the furnace is installed in a specially built enclosure occupying one half of a standard Spacelab experiment rack. Auxiliary equipment provides power, electronic control and data acquisition and cooling air to the furnace.

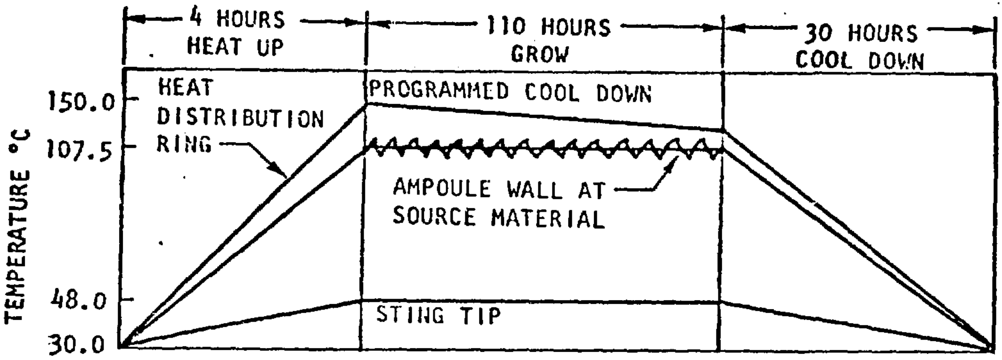

Fig. 3

The crew member subsequently checks out the control functions and initiates the experiment. The growth sequence is fully automated and consists of three essential parts: heat-up, growth and cool-down (see Figure 3). During the heat-up phase the temperatures at the different control points in the furnace (source, heat distribution ring, cooling sting) are increased in a controlled way to values which are optimal for growth. The growth phase is then started by gradually decreasing the pedestal temperature, primarily by reducing the heat input to the base heater. During the growth phase it is possible, when necessary, to oscillate the temperature of the source material so that periodic growing and etching of the crystal occurs to obtain more stable crystal growth and to avoid spurious nucleation. This is a technique usually employed during the early stages of the growth of mercuric iodide crystals [2] [3].

When the source material has almost been depleted or when the experiment time has elapsed, the system is gradually cooled down to ambient temperature and is returned to its storage container for re-entry.

During the growth sequence the crew member will be able to observe the crystal through a microscope/video system installed in the front wall of the experiment enclosure. The table on which the furnace is mounted can be rotated, so that all sides of the crystal are observable. The crew member will periodically monitor the growth and measure the dimensions of the crystal to determine the vapor transport rate which under the existing conditions is primarily governed by diffusion. When growth is not satisfactory the crew member can change the automatic sequencing program to establish more favorable conditions.

The observations of the crew member can be transmitted by voice link to personnel following the progress of the experiment on the ground (Payload Operations Control Center). During certain periods of the flight he will also be able to transmit real-time video images for on-ground observation of the experiment.

CRYSTAL EVALUATION

The characterization of the crystal grown in space needs careful planning since only a restricted amount of material will be available. There are basically two sets of experiments which one would like to perform on the material. The first group of experiments has the purpose of evaluating the structural quality of the crystal by direct measurement of absorption and scattering. The ...me piece of crystal can often be used for several types of experiments (e.g. ...ma-ray diffraction, luminescence, raman scattering, ESR). The second group ...xperiments is more directly related to the use of the material as a radi... detector. It involves the measurement of electrical charge carrier properties and requires processing of parts of the crystal into a device-type structure (sawing, polishing, contacting).

In the following we will summarily describe some of the evaluation experiments which can be expected to provide critical information about the quality of mercuric iodide crystals. The results obtained with the space-grown crystal will be compared with the results from ground-based crystals which have been grown in functionally equivalent furnaces.

Investigation of the crystal as received

After the crystal has been removed from the growth ampoule it will be photographed and be subjected to microscopic investigation. Since the material is transparent to red light (band gap 2.1 eV) it is often possible to observe structural defects of microscopic size or larger (inclusions, voids, cracks) by the way they scatter the light. The defects observed in this way can be mapped and will serve as a guide to determine which way the crystal will be divided in sections for further investigation.

Gamma-ray diffraction

Since mercuric iodide is a very good absorber for x-rays it is not possible to use low energy (10-40 keV) radiation to perform bulk transmission diffraction measurements. However, more penetrating radiation can be generated from artificial isotopes which in addition provide very monochromatic radiation. Energies in the range of 100-150 keV were used to perform rocking curve experiments on 1 mm thick samples of crystals. The widths of the diffraction peaks obtained is an indication of the structural quality of the material. Two types of extended defects have been observed which give rise to a mosaic substructure. Details of the techniques used and preliminary results have been published [4]. This method seems one of the most promising to obtain direct information regarding the structural quality of the crystal.

Luminescence

Measurements of the luminescence of mercuric iodide have been performed at 77°K and 4°K. The spectra obtained are very indicative of the electronic as well as structural quality of the material. Crystals from which high quality detectors can be fabricated have a very characteristic luminescence spectrum which increases in intensity as the structural quality of the material improves.

There are strong indications that luminescence measurements are sensitive to small stoichiometric changes in the material. Deviations from stoichiometry will create native defects which can contribute to the degradation of the electronic properties. Attempts have been made before to determine the stoichiometry of mercuric iodide by chemical methods, most recently by DeLong and Rosenberger [5], but the sensitivity of these methods seems to be limited to approximately 0.1 mole percent. The details of the luminescence measurements will be published shortly [6].

Mechanical measurements

The mosaic structure (polygonization) observed in even the best crystals makes one suspect that the crystal deforms during growth. To increase our understanding of the mechanical properties of the material, a program was started to measure its behavior under stress.

Mercuric iodide is a very soft material. The prevalent slip and cleavage plane is parallel to the c-plane. Compression measurements therefore were made of approximately 8 x 3 x 3 mm sections of crystal cut in such a way that the c-plane was at 45 degrees to the applied force. The measured values were in general so low that improvements in the sensitivity of the apparatus were repeatedly necessary. The lowest value of CRSS measured to date is 0.5 lb/sq.in. This implies that a crystal of mercuric iodide can support approximately 3 cm of its own material in a 1-g environment when loaded in the c-plane. Preliminary results indicate that the material becomes even softer at higher temperatures, but the data base has to be increased to be able to make conclusive statements.

The dislocation structure of mercuric iodide has been studied in detail by James and Milstein [7]. They come to the conclusion that sessile dislocations perpendicular to the c-plane hinder the movement of glide dislocations in the c-plane. Similar observations have been made by investigators in France [8].

Detector evaluation

To determine the quality of the crystal in terms of detector performance the response of the device to a standard radiation source is measured at different biases. The position of the spectral positions makes it possible to calculate the mobility-lifetime product of the charge carriers. Independent measurement of the risetime of the charge pulses generated by the radiation results in values for the lifetime, so that the mobility can then be calculated. Point-by-point evaluation of the detector response can be used to map inhomogeneities in the crystal. The device structure is also suitable for Thermally Stimulated Current (TSC) measurements by which it is possible to determine the energy levels and density of the electronic trapping centers.

SUMMARY AND CONCLUSIONS

A system has been developed to perform vapor crystal growth experiments in the environment of the Spacelab. Several methods have been described which can be used to evaluate the quality of the space-grown crystal. Some of these methods need further development to increase their sensitivity and reliability.

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