EXPERIMENT REQUIREMENTS AND IMPLEMENTATION PLAN (ERIP) FOR SEMICONDUCTOR MATERIALS GROWTH IN LOW-G ENVIRONMENT

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TABLE I.- Anticipated furnace performance of the MEA-A furnace for the growth of PbSnTe 10
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SUMMARY

The purpose of this experiment is to use the MEA-2 A facility to test the effect of the low gravity environment on suppressing convective mixing in the growth of Pb_{1-x}Sn_xTe crystals. This document describes the need to eliminate convection, the furnace characteristics and operation that will be required for successful experimental implementation, and to the level that is presently known, the measured physical properties of the Pb_{1-x}Sn_xTe system. In addition, a brief background of the present and potential utilization of Pb_{1-x}Sn_xTe is given. Additional experiments are anticipated in future MEA-A, improved MEA and other dedicated materials processing in space flight apparatus.
INTRODUCTION

Lead tin telluride is a narrow bandgap semiconductor material. The bandgap is adjustable from 0.21 eV to, theoretically, a zero bandgap by adjusting the ratio of the lead to tin on the metal side of the compound. This feature of PbSnTe makes it a desirable candidate for such applications as long wavelength infrared detectors and tunable diode lasers since these devices operate best when the wavelength of peak detectivity or emission is tailored to meet a specific need.

Both infrared detectors and tunable diode lasers have been built using PbSnTe but these devices have not yet been optimized for peak performance. The exact physics of device degradation due to crystalline defects is not complete; however, experience has helped to identify some of the interactions of defects and device characteristics.

Crystalline defects such as low angle grain boundaries and dislocations can produce diffusion paths which severely degrade diode junction characteristics and, hence, would be harmful to both detector and laser performance. Any compositional variations in the crystal will lower the peak detectivity of an infrared detector and can produce multimode emission from a laser. Strict control of the lattice dimension is necessary when the crystal is used as a substrate for epitaxial growth because a lattice mismatch produces strain in the crystal and this strain can be relieved by producing interfacial dislocations. Hence, the quality of the epitaxial device is limited by the quality of the starting material.

Since there is no commercial supplier of PbSnTe, device researchers and manufacturers must generally grow their own material. The three most common techniques of growth are:

1. Source ingot nucleation in which a polycrystalline aggregate is annealed for extended periods of time in an isothermal or nearly isothermal environment. Grain growth occurs during the anneal and small, highly faceted, low defect, crystals are produced.

2. Vapor transport in which PbSnTe vapor from a heated polycrystalline aggregate condenses on a slightly cooled, with respect to the source, substrate. Moderate sized crystals of moderate defect density are produced.

3. Liquid phase growth in which growth proceeds by controlled, directional solidification. This technique produces the largest crystals of the three techniques, but, to date, these crystals are the least homogeneous and exhibit the highest defect density.

Lead tin telluride single crystals can be grown on Earth but as previously noted, the best quality crystals are grown by the technique, ingot nucleation, which has the least interaction with the gravity field. The interaction of gravity on crystal growth is most evident in the liquid phase technique because the system, depending on growth orientation, will be either thermally or solutably unstable. The details of this instability will be discussed in a later section. The understanding of the interaction of gravity on crystal vapor growth is at a less mature state than the liquid growth but the present understanding centers around the establishment of a vapor
diffusion boundary layer of included gases that must be supplanted in order for growth to occur.

The experiments discussed in this report will attempt to determine the effects of gravity on the liquid phase growth techniques. With a better understanding of the growth mechanisms, the liquid techniques will be able to produce more nearly perfect crystals of PbSnTe.

LIST OF SYMBOLS

\( A_0 \)  
\[ \text{lattice constant, } 10^{-10} \text{ meters} \]

\( ^0C \)  
\[ \text{temperature, degrees Celsius} \]

\( C_L \)  
\[ \text{solutal concentration of liquid phase, mole fraction} \]

\( C_0 \)  
\[ \text{solutal concentration of starting material, mole fraction} \]

\( C_S \)  
\[ \text{solutal concentration of solid phase, mole fraction} \]

\( D \)  
\[ \text{solutal diffusion coefficient, } \text{cm}^2 \text{ sec}^{-1} \]

\( g \)  
\[ \text{acceleration due to body forces, cm sec}^{-2} \]

\( g_E \)  
\[ \text{acceleration due to gravity on Earth, } 981 \text{ cm sec}^{-2} \]

\( G \)  
\[ \text{temperature gradient, } ^0C \text{ cm}^{-1} \]

\( G_s \)  
\[ \text{solutal gradient, mole fraction cm}^{-1} \]

\( H \)  
\[ \text{dimension of furnace adiabatic zone, cm} \]

\( k \)  
\[ \text{segregation coefficient} \]

\( K_S \)  
\[ \text{thermal conductivity in solid, watts cm}^{-1} \text{ sec}^{-1} \]

\( K_L \)  
\[ \text{thermal conductivity in liquid, watts cm}^{-1} \text{ sec}^{-1} \]

\( L \)  
\[ \text{characteristic length, cm} \]

\( L \)  
\[ \text{latent heat of fusion, calories mole}^{-1} \]

\( m \)  
\[ \text{slope of liquidus line on phase diagram, } ^0C \text{ mole fraction}^{-1} \]

\( S \)  
\[ \text{solutal Rayleigh number} \]

\( N^{T}_{Ra} \)  
\[ \text{thermal Rayleigh number} \]

\( r \)  
\[ \text{radial dimension, cm} \]

\( R \)  
\[ \text{growth velocity, cm hr}^{-1} \]

\( t \)  
\[ \text{time,} \]
THE VALUE OF PROCESSING IN SPACE

The theory developed by Smith, Tiller, and Rutter (Ref. 1) and Tiller et al. (Ref. 2) which defines the condition for steady state growth with no macroscopic segregation clearly requires that the liquid transport be diffusion controlled only. A horizontal growth configuration will have thresholdless convection; hence, vertical growth in the gravity field is required.

The composition of \( Pb_{1-x}Sn_xTe \) which is of primary interest is \( (x) = 0.2 \). The liquid composition which will be in equilibrium with this solid is \( (x) = 0.29 \). Without considering, for the present, the effect of thermal dilatation, the density of liquid \( Pb_{.71}Sn_{.29}Te \) at the growth interface will be approximately 0.98 times as massive as the undisturbed liquid five diffusion lengths into the melt. The maximum SnTe compositional gradient will be at the growth interface and is given by:

\[
\frac{\partial C}{\partial Z} = - C \frac{1-k}{\alpha} \frac{R}{D}
\]

Assume a nominal growth rate of 4.5 mm/hr and a liquid diffusion coefficient of \( D = 7 \times 10^{-5} \text{ cm}^2/\text{sec} \) (Ref. 3), this gradient is

\[
\frac{\partial C}{\partial Z} = - 0.16 \text{ mole fraction SnTe/cm}
\]
The solutal Rayleigh number obtained with this concentration gradient and using a characteristic length of one centimeter is:

\[ N_{Ra}^S = \frac{\beta^*_x}{\nabla u} g L^4 \frac{\partial c}{\partial Z} \]  

(3)

\[ N_{Ra}^S = 2.1 \times 10^8 \]  

(4)

This extremely high Rayleigh number clearly exceeds any estimate of criticality if the growth direction is antiparallel to the gravity vector.

A vertical growth, parallel to the g-vector, will be in a thermally destabilizing configuration. Using the growth rate and diffusion coefficient used previously, a minimum thermal gradient of 15°C/cm at the growth interface, going into the liquid, is required to prevent constitutional supercooling. The Earth-based thermal Rayleigh number for this gradient and a 1 cm characteristic length is:

\[ N_{Ra}^T = \frac{\beta_T}{\alpha u} g L^4 \frac{\partial T}{\partial Z} \]  

(5)

\[ N_{Ra}^T = 1.9 \times 10^4 \]  

(6)

This Rayleigh number must be decreased by a factor greater than 100 to fall below the generally accepted critical values. The characteristic length term, L, can only be decreased at the expense of crystal size and a 1 cm size is already a minimum. The temperature growth rate can be decreased only by holding the ratio of temperature gradient and growth rate constant if constitutional supercooling is to be avoided. That is:

\[ \frac{1}{R} \frac{\partial T}{\partial Z} = \text{constant} \]  

(7)

Hence, a large drop in \( \partial T/\partial Z \) requires a large drop in \( R \) and a concomitant drop in the length of crystal growth needed to reach steady state which is given by

\[ Z_{ss} \propto \frac{D}{kR} \]  

(8)

Hence, due to these tradeoffs, a relatively high temperature gradient and growth rate are required and each of these parameters produce a driving force on convection.

The strength of these driving forces on the liquid is seen experimentally by examining the compositional profile of the solidified crystal. Figure 1 shows the distribution of SnTe in an Earth-grown PbSnTe crystal where the
average SnTe content was 20%. The solid line is the theoretical plot of the Scheil equation which is derived by assuming that the liquid was totally mixed.

It is the objective of this MEA A-2 experiment to test the extent that convection will be suppressed in the space environment. Future space flight experiments will build upon the results of the first flight. The details of this experiment will be discussed in the next section of this report.

**FLIGHT EXPERIMENT DEFINITION**

The experiment that will be conducted on the MEA A-2 is designed to test the ability of a PbSnTe directionally solidified crystal to obtain a steady-state compositional configuration in the low gravity environment offered by the Space Shuttle.

The PbSnTe will be contained in a sealed, fused quartz ampoule which, in turn, is packaged in an inconel cartridge provided by Marshall Space Flight Center. This package is shown in figure 2. The flight cartridge will contain two type K thermocouples which are also shown in figure 2 but the exact placement of these thermocouples is to be determined after more ground testing.

In the experiment, the entire contents of the ampoule will be melted and allowed to come to full thermal equilibrium at the maximum operating temperature of the MEA-A 2 furnace. The directional solidification of the contents will occur by programming the individual furnace elements down in temperature in a predetermined sequence to simulate the relative movement of the furnace with respect to the ampoule. After the entire contents of the ampoule are solidified the cooling rate can be increased to conserve the spacecraft battery supply.

**EXPERIMENT DEVELOPMENT**

This experiment is being developed to determine the extent that convective mixing in the PbSnTe growth system will be suppressed in the low gravity environment of the Space Shuttle. Theoretical and experimental data (Ref. 4) clearly show that PbSnTe is subjected to strong convection producing forces when grown on Earth. The capabilities of the MEA-A 2 are limited but are sufficient to grow a PbSnTe crystal that is large enough to see if diffusion controlled growth can be obtained. This chapter will describe the development of this experiment to date as well as the plans for further experiment development between now and flight delivery time.

The MEA-A furnace had to be tested to determine if a sufficient gradient can be obtained at the PbSnTe solidus temperature such that compositional steady state can be reached without either allowing a significant part of the diffusion tail to reach the end of the ampoule or allowing the interface to become unstable due to constitutional supercooling. Three equations must be considered to meet the above criteria. The first equation considered is for the composition of the solid in a diffusion controlled growth and is written as:

\[ C_s = C_L \left( 1 - \frac{T_s}{T_L} \right) \]

where:
- \( C_s \): Composition of the solid
- \( C_L \): Composition of the liquid
- \( T_s \): Temperature of the solid
- \( T_L \): Temperature of the liquid
This equation will specify the length of sample required to reach compositional steady state when a value for \( R \) is chosen and is valid only if the liquid region is large compared to the length of the diffusion barriers.

The second equation that must be considered describes the liquid diffusion front that is ahead of the solid-liquid interface. If compositional steady state has been reached, the equation of interest is:

\[
C_s(z^1) = C_o \left( 1 - \frac{1-k}{k} \exp\left( -\frac{Rz^1}{D} \right) \right) \text{ (Ref. 2)} \tag{9}
\]

where \( z^1 = 0 \) at the solid-liquid interface.

This equation determines the length, from the end of the boule, at which solute buildup at the end no longer can be ignored. That is, as the diffusion barrier hits the end of the ampoule the compositional gradient in the liquid ahead of the interface will decrease, hence, the solute concentration in the liquid and concomitantly in the solid, will increase. The equation that describes this final transient is given by Smith et al. (Ref. 1) as:

\[
C_s(z_2) = C_o \left[ 1 + \frac{\varphi(2n-1)}{\prod_{n=1}^{\infty} \frac{(1-k)(2-k)\cdots(n-k)}{(1+k)(2+k)\cdots(n+k)} \exp\left( -\frac{n(n+1)-1}{R} \frac{Rz_2}{D} \right) \right] \text{ (Ref. 2)} \tag{10}
\]

where \( z_2 \) the distance from the last to freeze end of the ampoule.

The third equation that must be considered is the criteria for interfacial stability given either by Tiller et al. (Ref. 2) or Mullins and Sekerka (Ref. 5). The Tiller equation is based on Rutter and Chalmer's (Ref. 6) supercooling theory and is given by:

\[
\frac{G}{R} > \frac{C_o}{D} \frac{1-k}{k} M \text{ (Ref. 2)} \tag{12}
\]

The Mullins-Sekerka equation is based on perturbation theory and is given by:

\[
\frac{G}{R} > \frac{C_o}{D} \frac{1-k}{k} M \frac{K_s + K_l}{2K_l} - \frac{\rho L}{2K_l} \text{ (Ref. 5)} \tag{13}
\]

The only processing variables in the above equations are the growth rate, \( R \), and the temperature gradient, \( G \). With the help of Jim Zweiner of MSFC, these variables have been measured with an instrumented boron nitride rod (see Fig. 3). It should be noted that the furnace and ampoule do not physically
move relative to each other hence crystal growth occurs by ramping the temperature of the furnace elements.

The temperature profiles in the rod at various times in a simulated growth cycle are shown in figure 4. This set of curves show a temperature gradient of approximately 20°C/cm at 890°C over the majority of the rod. The simulated growth rate is determined by measuring the rate at which the desired isotherm (i.e., the solidus temperature) can be translated through the ampoule. The resulting equation is:

\[ R = \frac{1}{G} \frac{\partial T}{\partial t} \bigg|_{T_s} \]  

The maximum possible growth rate is limited by the heat extraction capability of the furnace and has been measured as being greater than 9 cm/hr. The minimum growth rate will only be limited by the available computer capability and the temperature resolution of the control thermocouples.

The measured value of \( G (20^\circ C/cm) \) dictates a maximum allowable growth rate of 0.50 cm/hr if equation (12) is used and a rate of 0.68 cm/hr if equation (13) is used. The anticipated compositional profiles are plotted in figure 5 for each of these rates. Also in figure 5 is a plot of the profile if the liquid was totally mixed as has been observed in Earth grown crystals.

The difference in composition between the diffusion controlled growth and the totally mixed growth is quite sufficient to be observed with a standard wavelength dispersive electron microprobe analysis.

Radial temperature gradients will exist in the ampoule since heat must be coupled into and out of the ampoule. The radial gradients that are important are those that exist at the solid-liquid interface since these gradients will determine the shape of the interface. The shape and magnitude of the interface will change as growth proceeds because the gradient must be made to traverse the ampoule, hence, the position between heat insertion and heat extraction will change as a function of solidification length. That is, it is anticipated that the interface will be concave, looking from the liquid, in the initial solidification region and convex in the last to freeze region.

Measurements of the radial gradient are planned, however, the gradients have been calculated, using finite element analysis of the equation for the conservation of thermal energy, from the measured axial temperature and the measured values of the three thermocouples in the furnace walls. A plot of the calculated 890°C isotherms as the simulated growth proceeds from end to end in the ampoule is shown in figure 6.

The theory of Coriell and Sekerka (Ref. 7) and Carlson et al. (Ref. 8) has been used to calculate the solutal diffusion produced by these deviations of the interface from planarity. The projected compositional variations for the growth conditions are on the order of 1% due to the lack of interface planarity.
Additional tests are planned to better characterize the MEA-A furnace for growth of PbSnTe. One test will run an instrumented fused quartz rod in the inconel cartridge. Another test will run a PbSnTe charge in a fused quartz ampoule that has a capillary well in its axes for thermocouple insertion. These tests are designed to measure radial temperature gradients and to compare the measured temperature gradient in PbSnTe with that of the two inert materials that are the likely candidates of the flight development test cartridge. At the conclusion of these tests, an instrumented cartridge will be constructed and used to develop the software that will control the furnace temperatures during flight.

These tests must be conducted in a MEA-A furnace. Neither time nor resources permit the construction of such a system at the Langley Research Center, hence, these tests can only be conducted at the Marshall Space Flight Center. The importance of these tests cannot be over emphasized. If these tests cannot be conducted the flight experiment, as configured, would be meaningless.

FURNACE REQUIREMENTS FOR MEA EXPERIMENTS

The MEA-A 2 furnace will be used as it is. Preliminary tests have been conducted to test its adequacy and tests are continuing to optimize its operation for these experiments. Table I lists the anticipated performance of the furnace.
TABLE I.- Anticipated furnace performance of the MEA-A furnace for the growth of PbSnTe

1. Operating temperature range 400-1100°C
2. Ampoule size 20 mm OD
   60-80 mm long
3. Heated cavity control
   - Control setpoint stability ±1°C
   - Control setpoint accuracy ±5°C
   - Time to temperature >30 min
   - Linear gradient range >20°C/cm
   - Temperature overshoot <50°C
4. Cooling requirements
   0-10°C/min until T < 500°C then quench is OK
5. Operating environment Ar, N₂ or vacuum
6. Temperature measurements
   - Absolute accuracy ±1°C
   - Resolution ±1°C
   - Data rate 1 Sps
   - Number of sensors ≥ 2
7. Ampoule material Quartz in inconel cartridge
8. Special data
   - Residual acceleration measurements during growth
     - Magnitude <1 x 10⁻⁴  gₑ
     - Orientation 3 axes
DESCRIPTION OF EXPERIMENT SPECIMENS

A. Composition

52.3 wgt % lead
7.5 wgt % tin
40.2 wgt % tellurium

B. Size

20 mm OD, 60-80 mm long

C. Weight, approximate values

90 grams Pb\textsubscript{1-x}Sn\textsubscript{x}Te
20 grams Si\textsubscript{2}O\textsubscript{2} ampoule
18 grams cartridge

D. Materials

1. Sample Pb\textsubscript{x}Sn\textsubscript{1-x}Te
2. Ampoule
   Quartz in inconel cartridge
3. Thermocouple probes
   Type K in inconel sheaths

E. Materials Characteristics of PbTe, SnTe and Pb\textsubscript{x}Sn\textsubscript{1-x}Te

1. Thermal conductivity (Ref. 9)

\begin{tabular}{|c|c|}
\hline
\textbf{\(T(\degree C)\)} & \textbf{\(K \text{ (W cm}^{-1}\text{ K}^{-1})\)} \\
\hline
22 & 1.47 \times 10^{-2} \\
100 & 1.24 \times 10^{-2} \\
150 & 1.17 \times 10^{-2} \\
200 & 1.12 \times 10^{-2} \\
\hline
\end{tabular}
<table>
<thead>
<tr>
<th>Temperature (°C)</th>
<th>Solutal Diffusion Coefficient in Liquid (Ref. 3)</th>
</tr>
</thead>
<tbody>
<tr>
<td>220</td>
<td>$1.11 \times 10^{-2}$</td>
</tr>
<tr>
<td>300</td>
<td>$1.32 \times 10^{-2}$</td>
</tr>
<tr>
<td>400</td>
<td>$1.65 \times 10^{-2}$</td>
</tr>
<tr>
<td>500</td>
<td>$1.91 \times 10^{-2}$</td>
</tr>
<tr>
<td>600</td>
<td>$2.08 \times 10^{-2}$</td>
</tr>
<tr>
<td>700</td>
<td>$2.22 \times 10^{-2}$</td>
</tr>
<tr>
<td>750</td>
<td>$2.28 \times 10^{-2}$</td>
</tr>
</tbody>
</table>

Solutal Diffusion Coefficient in Liquid (Ref. 3)

PbTe, $D = 6 \times 10^{-5}$ cm$^2$/sec

SnTe, $D = 14 \times 10^{-5}$ cm$^2$/sec

$Pb(1-x)Sn_xTe, D = (3/7)(1-x) \times 1.4 \times 10^{-4}$ cm$^2$/sec
PbTe and SnTe - The tabulated data were taken from curves in reference 10.

<table>
<thead>
<tr>
<th>T(°C)</th>
<th>PbTe (K(\text{W cm}^{-1} \text{K}^{-1}))</th>
<th>SnTe (K(\text{W cm}^{-1} \text{K}^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>325</td>
<td>(x 10^{-2})</td>
<td>(2.0 \times 10^{-2})</td>
</tr>
<tr>
<td>425</td>
<td>(x 10^{-2})</td>
<td>(1.7 \times 10^{-2})</td>
</tr>
<tr>
<td>525</td>
<td>(1.0 \times 10^{-2})</td>
<td>(1.7 \times 10^{-2})</td>
</tr>
<tr>
<td>625</td>
<td>(1.2 \times 10^{-2})</td>
<td>(2.7 \times 10^{-2})</td>
</tr>
<tr>
<td>725</td>
<td>(1.3 \times 10^{-2})</td>
<td>(3.4 \times 10^{-2})</td>
</tr>
<tr>
<td>775</td>
<td>(1.7 \times 10^{-2})</td>
<td>(4.0 \times 10^{-2}) solid</td>
</tr>
<tr>
<td></td>
<td></td>
<td>(5.3 \times 10^{-2}) liquid</td>
</tr>
<tr>
<td>825</td>
<td>(2.0 \times 10^{-2})</td>
<td>(5.3 \times 10^{-2})</td>
</tr>
<tr>
<td>875</td>
<td>(2.7 \times 10^{-2})</td>
<td>(6.0 \times 10^{-2})</td>
</tr>
<tr>
<td>975</td>
<td>(4.5 \times 10^{-2}) solid</td>
<td>(7.3 \times 10^{-2})</td>
</tr>
<tr>
<td></td>
<td>(5.3 \times 10^{-2}) liquid</td>
<td></td>
</tr>
<tr>
<td>1075</td>
<td>(6.0 \times 10^{-2})</td>
<td>(8.3 \times 10^{-2})</td>
</tr>
<tr>
<td>1175</td>
<td>(7.2 \times 10^{-2})</td>
<td>(9.3 \times 10^{-2})</td>
</tr>
</tbody>
</table>

Specific Heat

\(\text{Pb}_{0.8}\text{Sn}_{0.2}\text{Te} \text{(Ref. 9)}\)

<table>
<thead>
<tr>
<th>T(°C)</th>
<th>(C_p \ (\text{W sec g}^{-1} \text{K}^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>22</td>
<td>0.1605</td>
</tr>
<tr>
<td>500</td>
<td>0.1720</td>
</tr>
</tbody>
</table>
Heat of Fusion (Ref. 11)

SnTe 10.8 K cal/mole
PbTe 13.8 K cal/mole

Density

The measured densities of solid and liquid PbTe and SnTe are easily fitted to a slope-intercept equation. That is:

\[ \rho = A_i + B_i T, \quad T \text{ is in } ^\circ C \]

The values of \( A \) and \( B \) are listed below along with the density at the melt temperature (Ref. 12):

<table>
<thead>
<tr>
<th>Material</th>
<th>A</th>
<th>B</th>
<th>( \rho ) at T melt</th>
</tr>
</thead>
<tbody>
<tr>
<td>PbTe solid</td>
<td>8.192</td>
<td>-5.38 \times 10^{-4}</td>
<td>7.693 gm/cm³</td>
</tr>
<tr>
<td>PbTe liquid</td>
<td>8.181</td>
<td>-8.43 \times 10^{-4}</td>
<td>7.40 gm/cm³</td>
</tr>
<tr>
<td>SnTe solid</td>
<td>6.467</td>
<td>-4.29 \times 10^{-4}</td>
<td>6.121 gm/cm³</td>
</tr>
<tr>
<td>SnTe liquid</td>
<td>6.514</td>
<td>-7.86 \times 10^{-4}</td>
<td>5.88 gm/cm³</td>
</tr>
</tbody>
</table>

The density of pseudobinary alloy of (1-X) mole fraction PbTe and X mole fraction SnTe is:
\[ \rho(x, T) = (1-x) \left( \frac{A_o \, (PbTe)}{A_o \, (Pb_{1-x}Sn_{x}Te)} \right)^{3} \rho_{PbTe} + \]

\[ x \left( \frac{A_o \, (SnTe)}{A_o \, (Pb_{1-x}Sn_{x}Te)} \right)^{3} \rho_{SnTe} \]

At the composition and temperature ranges of interest an error of less than 1 percent is incurred by using

\[ \rho(x, T) = (1-x) \rho_{PbTe}(T) + (x) \rho_{SnTe}(T) \]

**Coefficient of Bulk Thermal Expansion**

This parameter is defined as:
\[ \beta_T = \frac{1}{\nu} \frac{\partial \nu}{\partial T} \]

or

\[ \beta_T = \frac{-1}{\rho} \frac{\partial \rho}{\partial T} \]

which reduces to

\[ \beta_T = \frac{-1}{\rho(x,T)} (1-x) \beta_T(PbTe) + x \beta_T(SnTe) \]

for the alloy.

Note that \( \beta_T \) is a function of temperature as well as composition and phase, however the temperature dependence is small as compared to the other two variables.

Selected values are listed below:

<table>
<thead>
<tr>
<th>T</th>
<th>20(^\circ) C</th>
<th>Tm (solid)</th>
<th>Tm (liquid)</th>
<th>1150(^\circ) C</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \beta ) (PbTe)</td>
<td>65.7</td>
<td>69.9</td>
<td>113.9</td>
<td>116.9 ( \times 10^{-6}/\circ) C</td>
</tr>
<tr>
<td>( \beta ) (SnTe)</td>
<td>66.9</td>
<td>70.1</td>
<td>133.7</td>
<td>140.1 ( \times 10^{-6}/\circ) C</td>
</tr>
</tbody>
</table>
Coefficient of Bulk Solutal Expansion

This term is analogous to the thermal coefficient of expansion and is defined as:

$$\beta_x = \frac{1}{v} \frac{\partial v}{\partial x}$$

or

$$\beta_x = \frac{-1}{\rho} \frac{\partial \rho}{\partial x}$$

which reduces to

$$\beta_x(x, T) = \frac{\rho_{SnTe}(T) - \rho_{PbTe}(T)}{(1-x)\rho_{PbTe}(T) + x\rho_{SnTe}(T)}$$

for the alloy.

Selected values for Pb.8Sn.2 Te are:

**Solid (890°C)**

$$\beta_x = -0.22/\text{mole fraction of SnTe}$$

**Liquid (905°C)**

$$\beta_x = -0.23/\text{mole fraction of SnTe}$$
### Viscosity (Ref. 13)

<table>
<thead>
<tr>
<th>Temperature (°C)</th>
<th>PbTe</th>
<th>SnTe</th>
</tr>
</thead>
<tbody>
<tr>
<td>830</td>
<td></td>
<td>3.48 x 10^{-3}</td>
</tr>
<tr>
<td>865</td>
<td></td>
<td>3.26 x 10^{-3}</td>
</tr>
<tr>
<td>915</td>
<td></td>
<td>2.68 x 10^{-3}</td>
</tr>
<tr>
<td>940</td>
<td>2.43 x 10^{-3}</td>
<td></td>
</tr>
<tr>
<td>965</td>
<td></td>
<td>2.16 x 10^{-3}</td>
</tr>
<tr>
<td>970</td>
<td>2.16 x 10^{-3}</td>
<td></td>
</tr>
<tr>
<td>980</td>
<td>2.17 x 10^{-3}</td>
<td></td>
</tr>
<tr>
<td>1000</td>
<td>2.03 x 10^{-3}</td>
<td></td>
</tr>
<tr>
<td>1010</td>
<td></td>
<td>2.02 x 10^{-3}</td>
</tr>
<tr>
<td>1020</td>
<td>1.90 x 10^{-3}</td>
<td></td>
</tr>
<tr>
<td>1050</td>
<td></td>
<td>1.98 x 10^{-3}</td>
</tr>
<tr>
<td>1060</td>
<td>1.73 x 10^{-3}</td>
<td></td>
</tr>
<tr>
<td>1090</td>
<td>1.68 x 10^{-3}</td>
<td></td>
</tr>
<tr>
<td>1100</td>
<td>1.57 x 10^{-3}</td>
<td></td>
</tr>
<tr>
<td>1130</td>
<td></td>
<td>1.84 x 10^{-3}</td>
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<tr>
<td>1140</td>
<td>1.42 x 10^{-3}</td>
<td></td>
</tr>
<tr>
<td>1170</td>
<td></td>
<td>1.82 x 10^{-3}</td>
</tr>
<tr>
<td>1180</td>
<td>1.34 x 10^{-3}</td>
<td></td>
</tr>
<tr>
<td>1190</td>
<td>1.46 x 10^{-3}</td>
<td></td>
</tr>
<tr>
<td>1210</td>
<td></td>
<td>1.75 x 10^{-3}</td>
</tr>
<tr>
<td>1230</td>
<td>1.34 x 10^{-3}</td>
<td></td>
</tr>
</tbody>
</table>
SAFETY CONSIDERATIONS

The experiments described in this document present no risks of fire, explosion, high pressure glass breakage or, at room temperature, toxicity. However, both lead and tellurium emit toxic vapor at elevated temperatures. The accepted, long term exposure per cubic meter of air is 0.2 milligram for lead and 0.1 milligram for tellurium (Ref. 14). These levels exist at 500°C and 300°C, respectively, over the elements. However, PbTe does not dissociate in the vapor state and its vapor pressure is approximately equal to that of lead. Consequently, dangerous levels of PbTe will condense on surfaces which are cooler than 500°C.

FUTURE EFFORT

The space flight experiment on the MEA-A 2 is considered to be only the first of a series of experiments that will help establish the science and technology of directional solidification of compound semiconductor materials.
## MILESTONES

### Calendar

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<tbody>
<tr>
<td>1. ERIP</td>
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<td>2. Testing of inert samples</td>
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<td>3. Testing of PbSnTe gradient</td>
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<td>4. Delivery of flight samples</td>
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<td>5. Ground development tests</td>
<td>TBD</td>
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<td>6. GCEL tests</td>
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<td>7. MEA flight</td>
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<td>8. Analysis of flight sample</td>
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</tbody>
</table>
REFERENCES


Figure 1. Compositional profile of Earth grown crystal.

\[ C = C_0 (1 - \frac{1}{L})^{(k-1)} \]
Figure 2. Loaded flight cartridge.
Figure 3. Instrumented BN rod.
Figure 4. Temperature profiles in BN rod.
Figure 5. Calculated compositional profile
Figure 6. Plot of calculated 890° C isotherms in BN cartridge.
The purpose of this experiment is to use the MEA-2 A facility to test the effect of the low gravity environment on suppressing convective mixing in the growth of Pb$_{1-x}$Sn$_x$Te crystals. This document describes the need to eliminate convection, the furnace characteristics and operation that will be required for successful experimental implementation, and to the level that is presently known, the measured physical properties of the Pb$_{1-x}$Sn$_x$Te system. In addition, a brief background of the present and potential utilization of Pb$_{1-x}$Sn$_x$Te is given. Additional experiments are anticipated in future MEA-A, improved MEA and other dedicated materials processing in space flight apparatus.