Synthesis of layered tin monoselenide crystals doped with tellurium by Chemical Vapour Transport technique

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Abstract: Recently SnSe structure has been shown to posses remarkable best thermoelectric power performance. It will be worthwhile to see the effect of doping Tellurium in SnSe increases the thermoelectric power behavior of SnSe. With these things in mind we have grown SnSeTe crystals by Chemical Vapour Transport technique using Iodine as the transporting agent.

IndexTerms – Tin Selenide, Tellurium, Crystal growth, Vapour transport technique.

I. INTRODUCTION

To achieve the steady temperature gradient for the growth of SnSe and $SnSe_xTe_{1-x}$ (for x=0.9) crystals by chemical Vapour Transport technique using Iodine (I₂) as a transporting agent.

Layered Compounds with a layered structure, particularly the Group IV Metal Dichalcogenides[1] shows interesting optical and electrical properties. Selection of SnSe and its representative $SnSe_xTe_{1-x}$ (0<x<1) from orthorhombic IV- VI layered materials has been made because these compounds make them suitable candidates for applications in thermoelectric generators, detectors and emitters in the infra-red spectral region, as well as switching and memory devices.[2,3]

Several investigators[4-10] have taken up the growth of layered SnSe Crystals. In these cases, the crystals grew in the form of large single crystal. Thin platelets needed for the transport property measurements are, therefore, obtained from a large size crystal by the method of cleaving. The process of cleaving may introduce a large number of defects. It is, therefore, desirable to evolve a method by which the crystals can be grown in the form of thin platelets so that they can be used such for the measurements.

Growth of SnSe using iodine as a transporting agent has been reported by Domingo et al[11]. However, the exact details of the growth mechanism adopted by them are not available. Therefore, we took up the growth of SnSe by iodine as a transporting agent.

However, our interest got mainly focused on the Crystal Growth of SnSe mixed Crystals.

 $[SnS_2]_x$ - $[SnSe_2]_{1-x}$ layered crystals were grown using the Vertical Bridgman technique for various nominal compositions by M.M Gospodinov et al[12]. A systematic study on the growth and characterization of mixed single crystals with composition SnS_xSe_{1-x} (where x=0, 0.25, 0.50, 0.75 and 1), have been reported by T.H Patel et al. Growth, Synthesis, Structural, Photoelectrochemical and Dielectric studies of $SnSe_{0.5}Te_{0.5}$ single crystals using Vapour transport technique have been investigated by G.K.Solanki et al [13-14].

Thermoelectric materials can effectively convert waste heat to electricity, particularly in the prospects of energy and environment crisis. Therefore they have been extensively reviewed. Recently, Tin Selenide has shown a more promising thermoelectric material because of its ultralow thermal conductivity and high thermoelectric figure of merit[15-16].

Although, Te plays a dominant role in the preparation of high ZT thermoelectric material. it suffer from the disadvantage it is not abundant in Earth's crust. Therefore a lighter element Selenium, which has much higher earth abundance shows replace tellurium where posses.

Author has therefore worked mainly upon SnSe crystals which also have a good ZT value in order to have the advantage of Te for improvement of thermoelectric behavior; SnSe has been doped with traces of Tellurium.

II. EXPERIMENTAL

The powder compounds were prepared from the elements having the following purities (%) Sn 99.995%, Se 99.999% and Te 99.999%. Appropriate amounts of the powdered elements were introduced into a thoroughly cleaned quartz ampoule (2.2 cm inner diameter, 22 cm length) in stoichiometric proportions. A total charge of nearly 10 gm was used in each experiment. The ampoule containing the source material was evacuated to a pressure of 10^{-5} Torr. Then the ampoule containing the charge was stirred well for nearly 1 hour to ensure the proper mixing of the powdered elements. The homogeneous mixture was properly distributed along the length of the ampoule and it was placed into the furnace.

The temperature was slowly increased at a rate of 60K/hr. The temperature and the period for which the ampoule was kept in the furnace depend upon the material being grown. In the present case, the required temperature was 973 K. The ampoule was kept at this constant temperature for 3 days. After this period, the furnace was slowly cooled down to room temperature at the same rate 60 K/hr and then switched off. As a result fine free flowing; shiny homogenous polycrystalline material was achieved for each compound. The ampoule was broken and shaken well with help of agate mortar to prepare fine powder of this compound.

For actual growth process, the material was transferred into another thoroughly cleaned quartz ampoule. This ampoule with charge of material was evacuated at 10^{-5} Torr and then sealed. The ampoule was then inserted into a two-zone horizontal furnace. The front zone (reaction zone) of the furnace was maintained at 973 K while the back zone (growth zone) was kept at 923 K for the growth of $SnSe_{0.9}Te_{0.1}$ (I₂) single crystals.

III. RESULTS AND DISCUSSION

The growth of single crystals of any compound depends upon different parameters such as length of the ampoule, purity of the source materials used, quality of the quartz tube, achievement of proper vacuum, temperature distribution of the furnace, appropriate proportion of constituent element, time duration for crystal growth, availability of the continuous power i.e. electricity, the rate of increase and decrease of temperature etc.

The growth parameters for $SnSe_{0.9}Te_{0.1}$ (I₂) single crystals are shown in Table 1. The temperature was increased at the rate of 50K/hr, till it attained the required temperature in both the zones. For the growth of $SnSe_{0.9}Te_{0.1}$ crystals, the ampoule was left in the furnace for 170 Hr after that the temperature was decreased at the rate of 30 K /hr up to the room temperature. Then the furnace was switched off and the ampoule was carefully taken out from the furnace. The ampoule was finally broken and resulting crystals were collected.

| Crystal | Temperature distribution | | Growth Period |
|---|--------------------------|-----------------|---------------|
| | Reaction zone (K) | Growth zone (K) | (Hr) |
| SnSe _{0.9} Te _{0.1} (I ₂) | 973 | 923 | 170 |

The layered crystals of SnSe0.9Te0.1 have been successfully grown in the form of thin platelets by a Chemical Vapor Transport Technique using Iodine as a transporting agent is shown inside the Quartz Ampoule in Fig.1

Figure.1 Crystals of SnSe0.9Te0.1 (I2) inside the Quartz Ampoule.



Figure.2 Crystals of SnSe_{0.9}Te_{0.1} (I₂) in the form of thin platelets

IV. CONCLUSION

 $Crystals \ of \ SnSe_{0.9}Te_{0.1} \ have \ been \ successfully \ grown \ by \ Chemical \ Vapour \ Transport \ CVT \ technique \ using \ I_2 \ as \ transporting \ agent.$

Single Crystals of $SnSe_{0.9}Te_{0.1}$ were found to be grown in the form of thin platelets and needles. All of them showed a mirror-like metallic lustre.

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