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STUDY OF A HORIZONTAL GRADIENT FURNACE FOR VAPOUR GROWTH

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ABSTRACT

A horizontal linear gradient two zone furnace has been employed to grow single crystals of semiconducting compounds by physical vapour deposition (PVD) method. It was calibrated for various trials including, series and parallel combinations of coils, and set temperatures. The temperature profile of the furnace was studied by recording temperature versus distance along the axis of the furnace using chromel-alumel thermocouples. The results were analyzed systematically and the optimum conditions for setting source zone and growth zone temperature were identified.

Key words: *two zone furnace, physical vapor deposition, calibration, temperature profile.*

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1. Introduction

Crystals have been source of fascination and interest for mankind since he encountered with them. They have become an inevitable part of human life. Different methods are used to grow crystals. When it comes to the growth of very well defined crystals, the technique used for the growth is of much importance. The growth method determines the economic, qualitative and quantitative production of desired crystals. The knowledge of physical properties such as melting point, boiling point, thermal expansion coefficient of the material, density, reactivity with other materials etc., will be useful in determining growth methods and choosing crucibles. Apart from the purity of the material, the growth environment plays a significant role in growing technologically important crystals. The important specifications of the environment are temperature, atmosphere, container materials, parameters that control growth velocities, and growth instabilities. To grow single crystals of chalcogenide compounds, very high temperature and linear temperature gradient is required.

Furnaces are indispensable tools for solid state physicists. A variety of different kinds of furnaces exist, each with their own capabilities and limitations. Proper design of furnace is very much essential for providing the optimal condition for crystal growth. Various factors are to be considered while designing a furnace [1]. A thorough knowledge of the furnace demands the study of its behavior for different set temperatures and coil currents. The temperature variation along the length of the furnace gives the calibration graph which can be used to understand the temperature profile of the furnace. Horizontal vapour growth furnace made of glass is used to grow rubrene crystals [2]. Kunjomana and Chandrasekharan [3, 4] have described the vapour growth of needle shaped GaTe and Bi_2Te_3 crystals. Growth of SnS and SnSe platelet crystals has also been reported by physical vapour deposition method [5]. Large single crystals of high-vapour pressure opto-electronic materials such as the II-VI compounds have been grown using multi-tube vapour growth furnace in which the source and growth regions are two separate vertical furnaces connected by a horizontal transport passage [6]. For the preparation of vapour grown platelet shaped α-HgI_2 crystals horizontal configuration of the ampoule was found optimal [7]. The necessity of high temperature and cost effectiveness put together worked behind the indigenous fabrication of the horizontal vapour growth furnace. As such, the present report deals with the study and calibration of a two zone furnace suitable for the growth of crystals by vapour method.

2. Experimental

Crystals can be grown from vapours when the molecules of the gas attach themselves to a surface and move into the crystal arrangement [8]. In the PVD method, stoichiometric proportions of the elements are vacuum sealed in a quartz ampoule and placed inside the two zone furnace. The charge from the hot zone (source zone of the ampoule) vaporizes and is transported to the cold zone (growth zone of the ampoule) where it gets deposited yielding single crystals of desired composition. The coil windings of each zone are designed to achieve the desired source zone and growth zone temperatures. The horizontal vapour growth furnace has a 90 cm long ceramic tube with inner and outer diameters of 4.2 cm and 5.2 cm. Kanthal A1 wire of gauge 17 i.e. 1.42 mm thick and 1kg mass is wound over 50 cm. The coils are wound uniformly around the two zones, but the spacing between the turns is made different for both zones so as to maintain an appropriate temperature gradient. The resistances of growth zone and source zone are 34.1 Ω and 44.4 Ω respectively. Three leads A, B and C are drawn out from the two extreme ends and the middle junction respectively. Another ceramic tube (inner diameter 2.2 cm and outer diameter 3 cm) is inserted in the core of the furnace for ensuring the axial position of ampoule and thermocouple. The outer ceramic tube is wound around by zirconia wool blanket for high thermal insulation. A stainless steel chamber is provided as an outer covering. The furnace is supported by two iron stands. The temperature controlling mechanism consists of thermocouple, temperature controller and solid state relay.

3. Results and Discussion

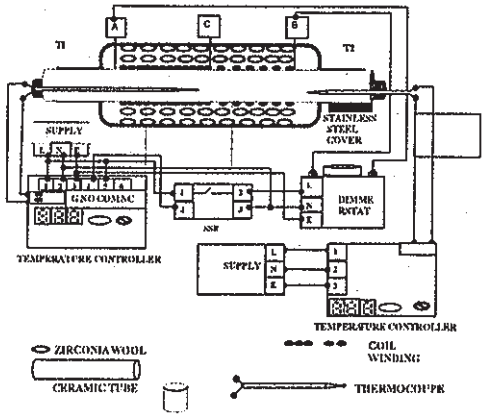

Fig. 1: Experimental arrangement for the study of temperature profile of the furnace.

The horizontal vapour growth furnace is calibrated for series/ parallel combinations of coils and different set temperatures. The whole experimental set up for calibration is shown in Fig. 1. An ac voltage of 225 V is supplied across the extreme terminals A and B. The furnace get heated due to resistive heating (I^2R) of the coil. The output of the chromel-alumel thermocouple, T1 (K-Type, 0.8 mm thick, 1 m long) is connected to the temperature controller (TC1). When the temperature exceeds the set temperature of TC1, solid state relay (SSR) is switched off and in turn dimmerstat is put off. As long as the furnace temperature exceeds the set temperature, the dimmerstat is maintained off. Once the temperature falls below the set temperature, SSR is switched on and the furnace gets heated up again. Hence a nearly constant temperature condition with a gradient is maintained inside the furnace.

Once the furnace was stabilized for the set temperature (With T1 at the maximum temperature point of the furnace) the furnace was calibrated using another chromel-alumel thermocouple, T2. Starting from one edge of the furnace, the temperature was noted for constant units of length. A plot between distance along the axis of furnace and temperature gave the calibration graph of the furnace for a particular set temperature. Different combination of connections were tried out to suit the required profile. Voltage was given across the terminals B and C (Low resistance region) and the temperature was recorded along the axis of the furnace. Though the heating was faster, a sharp hump appeared in the calibration graph (Fig. 2) due to the I^2 proportionality of power, I being different in the two coils. In the next attempt, voltage was given across the terminals A and B reducing the circuitry to two resistances connected in series. The calibration graph gave a flattened hump (Fig. 3). To ensure a better profile suitable for vapour growth, the variation of temperature with distance has also been studied by removing the inner ceramic tube. For keeping the thermocouple along the axis of the furnace, ceramic baffles were used. As a result, profile with fairly constant temperature region was obtained. The calibration graphs for set temperatures 600°C and 700°C were depicted in Figs. 4 and 5.

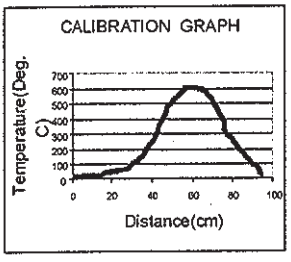

Fig. 2: 225V across B & C, set temperature 600°C with inner ceramic tube

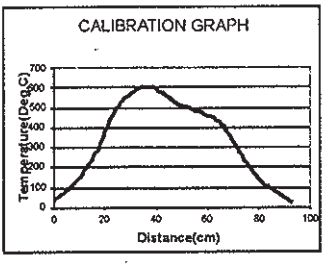

Fig. 3: 225V across A & B, set temperature 600°C, with inner ceramic tube

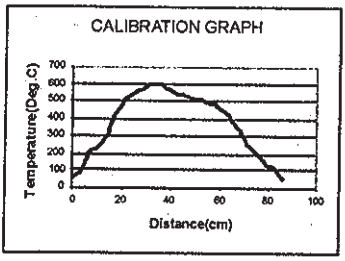

Fig. 4: 225V across A & B, set temperature 600°C

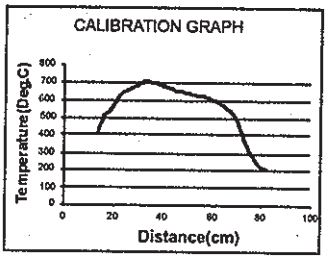

Fig. 5: 225V across A & B, set temperature 700°C

Thus the temperature profile without the inner ceramic tube was found to be better compared to the one with the inner tube. Also it provided the freedom of choice of ampoule diameter. Moreover, the heating and cooling rate was found to be lesser, which would favor defect free single crystal growth. Systematic trials were done for the set temperature 700°C, for which a flat profile with a linear region of 20 cm was obtained. The result was found to be optimum for the growth of single crystals by vapor growth method.

4. Conclusion

The horizontal vapour growth furnace is calibrated for different furnace coil combinations and set temperatures. Systematic trials gave temperature profile with a linear temperature gradient. The profile has an isothermal region of length 20cm. The results obtained are suitable for crystal growth using physical vapour deposition method.

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