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A Novel System for Growth of Single Crystals from the Melt with an Innovative New Pulling Mechanism

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This paper describes a crystal growth system employing novel concepts in fabrication of a high temperature furnace, thermocouples and a novel crystal pulling mechanism. This has reduced the costs substantially, without compromising the quality of grown crystals. The core of the furnace, a wooden cylindrical dummy had been prepared with equi-spaced helical groves with widths equaling the diameter of the heating wire on its outer surface machined by a lathe machine. The Kanthal heating wire was wound in the groves. It was covered with a thick layer of natural clay available locally. After the clay had dried up, an electric current was passed through the heating wire and the wooden frame was burnt out. A thick layer of the clay was applied on the inner and the outer surfaces. The furnace can operate at temperatures up to about 1000 °C. The temperature was measured with a chromel-alumel thermocouple prepared by an ingenious spot-welding technique established in the laboratory. The seed holder was hanged above the melt kept in the crucible with help of a float kept in a water container, which has a small tap at the bottom. When the tap is opened the float goes down and the seed assembly goes up. In this manner a quality pulling system, without any motor has been developed and reported here. It has been possible to grow good quality crystals of potassium chloride with excellent diameter control.

Keywords: Novel system for melt growth of crystals, New pulling mechanism, Fabrication of a high temperature furnace with local clay, Growth of potassium chloride crystal

1 Introduction

Single crystals are extensively being used for research and development as well as in numerous applications including the mass production of advance devices like microprocessors and some of the devices used in systems employing artificial intelligence and machine language and even in parts of the quantum computers. Indeed, these are the starting materials for fabrication of numerous devices like computer chips, detectors and systems used in bio-medical field also. Single crystals are produced from melts[1-8] solutions as well as from the vapour phase. The crystals for technological applications are required to fulfill stringent specifications in terms of their compositions and purity as well as crystallographic structure and lattice imperfections to ensure that the properties required for the particular applications are achieved repeatedly[9]. For scientific understanding of properties of materials also one requires single crystals that are prepared under controlled well-defined conditions. Special efforts are made to ensure that desired composition and near absence of unwanted impurities as well as prescribed low density of defects are achieved. Such crystals are prepared for research as well as for industrial applications. Of course, the desired impurities for different applications are introduced in a controlled manner. Also, critical growth conditions, particularly the temperature uniformity around the growing crystals are essential in ensuring a low level of crystal defects.[10-11]

Single crystals are grown from the solutions, from the melt as well as from the vapour phase. Melt growth of crystals is the most commonly used technique for research as well as for advanced applications. The following techniques are commonly employed for melt growth of crystals: the Czochralski method, the Kyropolous tecnique and the float zone crystal growth systems. Among these, the most commonly used technique is the Czochralski technique. Besides R&D application, this technique is extensively employed in industrial production of very large diameter crystals of silicon for fabrication of microelectronics and other devices.

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In the Czochralski technique the starting material is generally in the form of a powder. It is filled in a crucible, which can operate at temperatures above the melting point of the material (*e.g.* 1414 °C for silicon and 770 °C for potassium chloride). It should be inert and should not react with the material used for crystal growth. The crucible is placed inside a furnace, which can operate at temperatures above the melting point of the material. The temperature in the space around the crucible is kept uniform by proper care taken in the fabrication of the furnace. After melting the powder, the temperature of the furnace is maintained just above the melting point of the material. A small piece of a single crystal of the material to be grown is used as a seed. The seed is held in a seed holder prepared from a material that has a higher melting point and does not react with it. The seed is dipped slightly in the melt. This leads to the exchange of heat between the seed and its holder and the melt. It is to be ensured that the melt temperature is such that the seed neither melts away when it comes in touch with the melt neither it freezes the melt on coming in touch with the same by extracting too much heat from the melt. After the equilibrium is attained, the seed is slowly pulled and the liquid around it freezes leading to the nucleation of the new crystal. The nucleus is pulled continuously, and the temperature of the melt is reduced gradually in a controlled manner, leading to an increase in the diameter of the growing crystal. This process is continued till the final desired diameter of the crystal is attained. It is then pulled continuously, while maintaining the temperature constant. However, while starting the crystal growth on a new facility, one has no seed to begin with. To overcome this difficulty, a platinum wire is dipped in the melt and it is pulled slightly and a drop of the melt at the end of the wire becomes a small nucleus. A picture of a crystal of potassium chloride grown in this manner is shown later.

For establishing facilities for crystal growth, the following key steps have to be taken:

(i) fabrication or purchase of an electric furnace, which gives a uniform temperature in the desired range in a volume greater than the volume of the crucible, which is several centimeters in diameter as well as in length;

(ii) establishing a reliable temperature measurement system, which allows control of temperature in the desired range; and

(iii) establishing a facility for continuously pulling the growing crystal till the desired dimensions of the crystal are attained.

Often elaborate sophisticated equipment are employed for growth of good quality crystals, which take care of all the three requirements listed above.

In this paper, we describe a crystal growth system, which employs a few novel low-cost approaches to enable preparation of good quality crystals. Particular emphasis has been given to fabrication of the system in-house and usage of local materials like clays as the insulation materials. The thermocouples used in these experiments were also prepared in the laboratory, which served as good training point for the students. Also, the pulling of the crystal has been accomplished by an un-precedented ingenious new low-cost system, which does not use any electric motor. The quality of this system has been demonstrated by examples of the growth of good quality potassium chloride crystals.

2 Growth of Single Crystals from the Melt based on the New Concepts

2.1 The crystal growth system

Fig 1 shows a schematic diagram of the new Crystal Growth System. It consists of the following constituents:

(i) A cylindrical furnace F;

(ii) A crucible C placed on a ceramic support S;

(iii) A thermocouple Th for monitoring the temperature in close proximity of the growing crystal;

(iv) A seed-holder SH, in which a small piece of single crystal or a platinum wire or wire of other inert material is fixed;

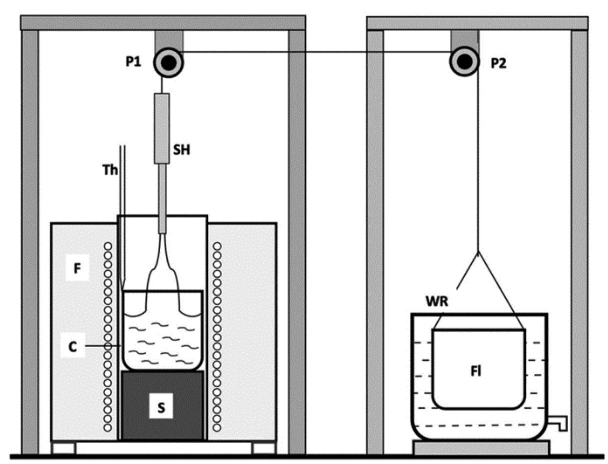

Fig. 1 — A schematic diagram of the equipment developed for growth of good quality crystals from the melt by the Czochralski method (F - cylindrical furnace, C - crucible, S - ceramic support, Th - thermocouple, SH - seed-holder, P1 - pulley P1, P2 - pulley, F - float F, WR - water reservoir, T - water tap)

(v) Two pulleys P1 and P2, the seed is hanged from pulley P1, whose other end is attached to a float F via pulley P2;

(vi) F is freely floating on the surface of the water reservoir WR; and

(vii) A water tap T attached to the bottom of the reservoir.

It may be mentioned that to begin with seed crystals can be grown by using thin platinum wires as seeds.

The fabrication of the thermocouples, the electric furnace and the crystal pulling system have been described separately in the following.

2.3 Fabrication of the thermocouples

The joints between two dissimilar metal wires, preferable prepared by spot welding are a very reliable probes to measure temperatures. As the temperature of the substance in contact with the joint increases or decreases a small voltage, generally in millivolts, is produced between the two wires of the thermocouple. Standard tables (For example, Type K Thermocouple: Temperature *vs* Milivolt Table[12]) are available in the literature, which give the temperature corresponding to potential difference between the wires of the thermocouple. This voltage can be conveniently measured by a millivotmeter. Most of the multimeters have a provision to measure the small voltages in millivolts.

A graphite rod of about 10 mm diameter and about 150 mm length, a usual/nose plier and a variac are the basic components of this system. One end of the carbon rod is connected to one terminal of the variac with a normal flexible conducting multi-strand electric wire. One end of another piece of the similar wire was attached to one arm of the plier, whose other end is connected to the free terminal of the variac. The movable arms of the plier are covered with an insulating material layer. The wires to be spot welded are put together and twisted so that their joint becomes small point-like feature. It is held tightly in the plier. It is momentarily brought in close contact with the carbon rod. This leads to the spot welding of the joint. The two wires are insulated from each other by an insulating ceramic sleeve with two holes. When the spot-welded joint is brought in touch with a heated substance, which may be a solid, liquid or gaseous a potential difference develops between the wires. From the measured potential difference temperature of the object can be determined with the help of standard tables available (referred to above).

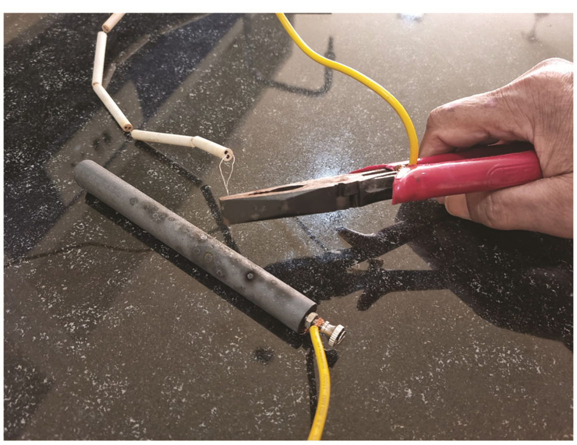

Fig. 2 — A photograph depicting preparation of a thermocouple by an ingenious technique of spot welding.

We had used chromel-alumel thermocouples in this investigation. These can easily function in the temperature range: -200 °C to 1350 °C. The same approach can be used to prepare thermocouples of copper-constantan or platinum-platinum rhodium wires or other wires.

2.4 The Fabrication of the High Temperature Furnace

The furnace has been designed to operate at temperatures of up to about 1000 °C. The diameter of the heated zone was kept as ~75 mm, so that crucibles of diameters of ~ 50-65 mm could be used for the crystal growth experiments. A thin Kanthal wire (~ 15 gauge) has been used as the heating element. The height of the furnace has been selected as 200 mm.

To begin with a solid wooden cylinder of diameter 110 mm and length 200 mm was prepared on a lathe. This cylinder was machined to convert it into the shape shown in Fig. 3. There are equidistant helical

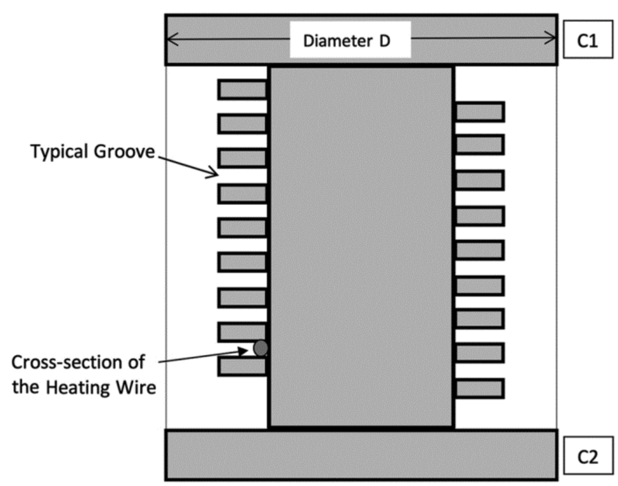

Fig. 3 — A schematic diagram showing the wooden frame used for fabrication of the furnace with uniform temperature

grooves along the entire length of this cylinder. The dimensions of these are groves are equal to the diameter of the heating wire of Kanthal. The Kanthal wire is tightly wound around the wooden cylinder keeping them inside the groves. In the end, two long ends of the wire are left free, to be connected to a power supply, as will be described later. This process ensures a very uniform distance between the adjoining segments of the heating wires, and therefore ensuring the uniformity of the temperature in the furnace.

The next step is to cover the wooden cylinder with the heating wires wound around it with a thick paste of clay. The paste is prepared by mixing the dry clay with water. The upper and lower collars C1 and C2 (Fig. 3) serve as sharp boundaries between the clay and the wooden frame at the bottom as well as at the top.

After the clay had dried, the ends of the heating wire are connected to a Variac, which is energized through a voltage stabilizer. The electric current in the furnace is gradually increased till it leads to the burning of the entire wooden frame. This process leads to the inner surface consisting of exposed heating wires. A thin layer of clay is pasted on these and allowed to dry up. The basic part of the furnace is complete at this stage. It is placed inside a metal frame F and ends of the heating wires are taken out and connected to a small insulating sheet fixed with nuts and bolts, which enable connection of the furnace to a Variac. The space between the frame walls and the outer surface of furnace is filled with dry clay, which serves as the insulator. Fig. 4 shows a schematic diagram of the assembled furnace. The vertical as well as horizontal gradients in the furnace were ensured to be low.

2.5 The crystal pulling mechanism

Several varieties of pulling systems have been developed from time to time for growth of crystals from the melt. A small piece of the crystal of the material to be grown as a single crystal is used as a seed. The seed is fixed at the bottom of a metallic rod. The melt of the material to be grown is maintained at a temperature slightly above its melting point. The seed crystal is slowly dipped in the melt and the temperature is maintained in such a manner that it remains in equilibrium without significant change in its diameter. At this stage the pulling of the seed is started and the temperature is reduced slowly to increase the diameter of the growing crystal. Often necking is introduced to ensure low density of defects.

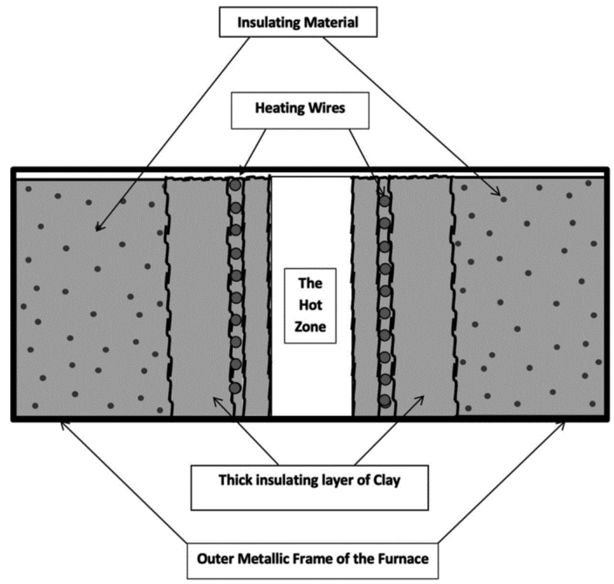

Fig. 4 — A schematic diagram of the furnace developed for crystal growth experiments. It shows 'the hot zone', 'the heating wires' and 'insulating material'

The pulling is continued after the desired diameter of the crystal has been achieved. Normally, electric motors, capable of providing variable speeds with suitable gear systems are employed to pull the crystals during growth from the melt. We report here a new ingenious mechanism, in which no electric motor is employed.

The basic principle of the new mechanism is quite simple. We have used a water container, which has a water tap fixed near its bottom, as shown in Fig. 1. When the water is filled in the container and the tap is opened, the water level in the container will keep on going down. The rate of fall of the water level depends on the opening of the tap, which is controllable. To make use of it in crystal growth experiment, a float of reasonable weight is allowed to float on top of the water. This is connected to the growing crystal with the help of two pulleys as, shown in the figure. The drainage from the tap is adjusted to obtain the desired pulling rate. Fig. 1 shows a schematic diagram of a system developed based on this principle and used for the melt growth of crystals.

3 Some Typical Results

The crystal growth system described above has been employed to grow good quality single crystals of potassium chloride. An alumina crucible of approximately 60 mm diameter was employed. It rested on a cylindrical clay spacer of diameter ~55

Table 1 — Comparison of the traditional crystal growth techniques and the new approach reported in this paper.

S. No.	Traditional Technique	New approach reported in this paper
1	Commercial furnaces comprise of a ceramic tube around which the heating wire is wound in an approximate equidistance manner	We have developed a technique which enables equidistant heating wires leading to high-level uniformity in temperature distribution.
2	The insulating material between the cover of the furnace and the heating tube is alumina or similar powders	We have utilized clays from local sources. Of course, the particle size and uniformity of clay was ensured
3	The crystals are pulled from the melt by using a suitable gear system coupled to an electric motor or a variable speed motor	We had coupled the growing crystal to a float, freely floating in a water reservoir, which had a tap to release water in a controlled manner to achieve the desired rate of growth
4	Commercially available thermocouples are used, which are enclosed in a thick ceramic tube	We had prepared thermocouples by a new spot-welding technique, which enable us to reduce the diameter of the same.

Fig. 5 — A photograph of a potassium chloride crystal grown on the newly developed system

mm and height of ~70 mm. The electrical power to the furnace was provided by a Variac, whose power source was a voltage stabilizer. To begin with we utilized a thin platinum wire fixed at the bottom of the seed holder to prepare a small seed. It was slightly dipped in the melt and slowly pulled up. A small drop of the melt, in this case potassium chloride was allowed to solidify into a small crystal. At this stage, the pulling is optimized by adjusting the rate of release of water from the container through the tap attached at the bottom. The temperature of the melt is decreased in a controlled manner till the size of the growing crystal reaches the desired dimensions. Thereafter, the growth is continued till the desired length of the growing crystal is realized. Fig. 5 is a photograph of a typical potassium chloride crystal grown with a platinum wire used for initiating the nucleation of the seed crystal. This platinum wire is seen in the picture. The crystal has an approximate diameter of 10 mm and a length of about 80 mm. Crystals like this can become a source of seed crystals or platelets for different applications that are cleaved from it. In the next phase, we have grown potassium chloride crystals by employing single crystal seeds.

A comparison of the traditional methods and the techniques reported in this paper is given in Table 1.

4 Conclusions

In this paper we have described details of fabrication of a crystal growth system comprising of a high temperature furnace capable of operating at stable temperatures of up to 1000 °C. The temperatures in the furnace were monitored by chromel-alumel thermocouples fabricated in the laboratory by an innovative spot-welding technique. A novel pulling system has been developed which does not require any electric motor. The uniform diameter of the potassium chloride crystal grown on the system amply demonstrates the quality of the temperature control as well as the new low cost pulling method.

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