APPARATUS FOR GROWING SINGLE CRYSTALS
AND PURIFYING SUBSTANCES

Filed Dec. 30, 1952

Fig. 1

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AGENT
APPARATUS FOR GROWING SINGLE CRYSTALS AND PURIFYING SUBSTANCES
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Application December 30, 1952, Serial No. 328,833
4 Claims. (Cl. 23—273)
(Granted under Title 35, U. S. Code (1952), sec. 266)

The invention described herein may be manufactured and used by or for the Government of the United States for governmental purposes without the payment to me of any royalty thereon in accordance with the provisions of the act of March 3, 1883, as amended (45 Stat. 467; 35 U. S. C. 45).

The present invention relates to an apparatus for growing single crystals and for purifying substances, and in particular to an apparatus which provides a plane temperature boundary at the interface of two immiscible liquids which are maintained at different temperatures.

It was well known in the prior art that single crystals of a substance could be produced by passing the substance which was to be formed into a crystal from a first temperature zone above the melting point of the substance through a plane temperature boundary into a second temperature zone below the melting point of the substance under consideration. It was also well known that as growth of the crystal proceeded the substance was purified by rejection of less pure material. One of the prior art methods of producing this plane temperature boundary between two different temperature zones consisted of two combustion furnaces separated by a thin layer of a heat insulating material which regulated the temperature boundary between the two zones. The heat insulating layer had an opening which nearly adhered to the walls of the crystallization tube containing the substance to be crystallized, which tube was withdrawn from the upper to the lower furnace by means of a platform elevator. There are several disadvantages to this method, such as, an inferior temperature boundary and nonuniform temperatures in the heated zones due to circulation of air between the two furnaces. This latter disadvantage was particularly serious in that it caused a diffuse zone of temperature on either side of the boundary which produced a horizontal temperature gradient. This often made for the nonuniform horizontal formation of the crystals and thereby caused stresses in the final product. Also, there is a large time lag in obtaining equilibrium temperatures, which is due to the low heat capacity of the air in the system. In this prior art apparatus it was not possible to observe the process during the growth of the crystal, and this also made it impossible to remelt a selected portion of the sample to remove the rejected impurities.

The object of the present invention is to provide an apparatus for growing single crystals in which temperature equilibrium may be quickly established and easily maintained.

Another object of the invention is to provide an apparatus for growing single crystals in which uniform temperatures may be maintained in the heated zones.

Another object of the invention is to provide an apparatus for growing single crystals in which there is a nonuniform zone of temperature from the temperature boundary.

Another object of the invention is to provide an apparatus for growing single crystals in which the temperatures in the two zones may be varied over wide ranges without appreciably affecting the quality of the temperature boundary.

Another object of the invention is to provide an apparatus for growing single crystals in which the passage of the crystallization tube from one zone to the other does not appreciably affect the temperature boundary.

Another object of the invention is to provide an apparatus in which selected portions of the material may be remelted as desired.

Another object of the invention is to provide between the two temperature zones a stable plane temperature boundary having a very high temperature gradient.

Another object of the invention is to provide a system in which there is a minimum of heat exchange between the two temperature zones.

In accordance with the present invention there is provided a column which contains two immiscible liquids heated to different temperatures thereby producing two uniform temperature zones and a plane temperature boundary at their interface. Resistance wire is wound around the two sections of the column to heat the respective liquids to the desired different temperatures. The level of the interface in the column is automatically maintained at a predetermined height. A motor is used to lower the crystallization tube through the temperature interface at a constant rate. The crystallization tube is suspended from a wire and is weighted and centered by a weight attached to the bottom.

Other uses and advantages of the invention will become apparent upon reference to the specification and drawings.

Figure 1 is a drawing of the overall apparatus of the present invention.

Figure 2 is a detailed view of the crystallization tube.

Figure 3 is a drawing of a modification of the apparatus showing re-entrant thermal wells for more accurately measuring the temperatures in the two zones.

Referring to Figure 1, there is shown a glass column containing two immiscible liquids. The first immiscible liquid is contained in the upper portion of the glass column above the plane temperature boundary which is represented by the dotted line. The second immiscible liquid is located below this plane temperature boundary. The column is provided with an arm 3 which permits overflow of the upper liquid into the recycling reservoir 4. The glass tube 6 is inserted into the reservoir and connected to the check valve 7. The check valve 7 is connected to another check valve 8 by the tubing 9. Connected into the tubing 9 between the two check valves is another tube 11 which is connected to a pump 12 which consists of a piston 13 riding in a cylinder 14. The piston is operated by a motor 16. The liquid flowing through the check valve 8 is fed back into the column 1 through the tubing 17 and cold water condenser 18. The bottom end of the cold water condenser passes through a hole in the stopper 19 in the top of the column 1. It will be noted that the tube 17 is offset from the center of the cold water condenser so that the cable 21 which supports the crystallization tube 22 may pass through the center of the cooler column of the cold water condenser. The cable 21 is attached to an essentially constant speed motor 23, which motor controls the speed at which the crystallization tube is lowered through the column 1. Suspending from the bottom of the crystallization tube 22 is the drive weight and guide 24 which is used to maintain the crystallization tube erect and to keep the cable 21 taut. The bottom of the column 1 is connected through the U-shaped tube 26 and the rubber tubing 27 to the leveling bulb 28. The arm 29 of the leveling bulb directs the overflow from the bulb into the reservoir 31. The upper portion of the column 1 above the plane temperature boundary 2 is wound
with resistance wire 30 which is connected to a variable source of voltage, not shown, for maintaining the upper liquid at a predetermined temperature. The lower portion of the column below the plane temperature boundary is also wound with resistance wire 35 which is used to maintain this portion of the column at a predetermined temperature.

In operation the two immiscible liquid layers are brought to the desired temperature above and below the freezing point of the substance in question. The interface between the two liquids is adjusted to the desired height in the column by lowering or raising the leveling bulb 28. The sample, previously sealed in the crystallization tube 23, is saturated with a vapor pressure, is attached to the leveling mechanism and the recycling system for the upper layer is started. The combination of the leveling bulb 28 and the recirculation system maintains the temperature boundary at a constant height during the operation. There are many other well known methods of keeping the boundary at a constant height and the present method forms no part of the present invention. In some instances it may be desirable to allow the level to rise or fall in which case the heating elements are raised or lowered also.

The sample in the crystallization tube is allowed to melt completely and then lowered to a position in the column so that the bottom portion of the tube 22, which has a capillary tube, is just above the liquid interface. The motor 23 is started and the crystallization tube is slowly lowered through the temperature interface. In some cases it might be advantageous to reverse the temperatures of the two zones and to raise the tube from the lower to the upper level. After complete crystallization (that is, when the sample is in the lower liquid) the sample is slowly brought to room temperature to prevent fractures of the crystal after growth.

The apparatus provides many advantages over that of the prior art, since it is possible to maintain a very sharp temperature boundary between the two immiscible liquids and to maintain nondiffuse temperature zones on either side of the interface. This apparatus is very flexible in that the temperature of each immiscible layer can be independently varied over very wide limits and still permit the maintenance of a good temperature boundary condition at the interface. This was one of the main disadvantages of the prior art system where there was continuity between the air in the two furnaces which readily permitted conduction of heat from one to the other. The advantages stem from the fact that the heat contact within each liquid is excellent while heat contact between the two different liquids is very poor. Also the present apparatus provides for the passage of the crystallization tube from one temperature zone to the other without destroying the boundary condition. This is made possible by using immiscible liquids which will repel one another, and therefore permits a minimum of intermingling and heat exchange. Of very great importance is the fact that the sharp temperature boundary permits remelting of a selected portion of the crystal, which is very useful in separating the crystal from the rejected impurities.

Figure 2 shows a cross section of the crystallization tube which is used in the practice of the present invention. The crystallization tube has the general shape of an elongated hexagon to which is connected a capillary tube 32 having a wall thickness, from which the drive winding and guide are attached. To the upper end of the hexagon is attached a column 33 that is necked down and then formed into a series of bolts connected by necked-down portions. The purpose for this is as follows: It is apparent that during crystallization and/or purification of a substance by the method just described, the impurities in the material initially placed in the crystallization tube 23 will be expelled during the formation of a crystal and will rise to the top of the mass. These impurities will gather in the V1 region. Thereafter, the V3 portion is remelted, using a reverse heating system in which the lower liquid is at the higher temperature, and suspending the tube upside down in the column. The very sharp plane temperature boundary makes it possible to select with great accuracy the exact portion which will be remelted. The material that had been in the V3 section will now run down into the lowermost portion of the tube Vv and, after solidification, this part can be sealed off from the rest of the system and removed. Then the sample tube minus this lowermost section is reintroduced into the system, the remaining impurities being as before. The rejected impurities from the second run reside in the portion V2 and are removed in the manner just described. This can be carried on for as long as it is practical or desirable, depending upon the degree of purity required.

Figure 3 shows a cross section of another embodiment of the invention showing only the main column. In this modification the center portion 36 of the column 1 is increased in diameter to allow the re-entrant tubes 37 to be formed in the column. These tubes permit the temperature in the column to be measured at various points to insure a uniform temperature throughout the upper and lower liquids, respectively, before the system is placed in operation.

It will be apparent that the embodiments shown are only exemplary and that various modifications can be made in construction and arrangement within the scope of my invention as defined in the appended claims.

I claim:

1. An apparatus for growing crystals of a substance, comprising a hollow vertical shell, means for maintaining a column of liquids in said shell, two immiscible liquids in said shell constituting said column of liquids producing a plane temperature boundary at the interface of said liquids, means for separately heating said liquids to different temperatures, respectively, one of said liquids being heated above the melting point of the substance to be crystallized and the second of said liquids being heated to a point below the melting point of the substance, a crystallization tube positioned within said shell and immersed in said liquids for holding the substance to be crystallized and means for withdrawing said tube from the first liquid into the second liquid.

2. The invention of claim 1 in which said crystallization tube is provided with at least one impurity collection chamber having a neck portion adapted to be sealed off to enable removal of said collection chamber from the crystallization tube.

3. An apparatus for growing crystals of a substance, comprising a hollow vertical shell, means for maintaining a column of liquids in said shell, two immiscible liquids in said shell constituting said column of liquids producing a plane temperature boundary at the interface of said liquids, means for separately heating said liquids to different temperatures, respectively, one of said liquids being heated above the melting point of the substance to be crystallized and the second of said liquids being heated to a point below the melting point of the substance, a crystallization tube positioned within said shell and immersed in said liquids for holding the substance to be crystallized and means for withdrawing said tube from the first liquid into the second liquid at a predetermined rate, and means for maintaining the interface at a predetermined height.

4. An apparatus for growing single crystals of a substance comprising a first container, a first and a second liquid located in said container, said liquids being immiscible and producing a plane boundary at the interface of said liquids, means for maintaining said first liquid above the melting point of the substance to be crystallized and means for maintaining said second liquid below the melting point of said substance, a second crystallization container in which said substance is located.
positioned within said first container and immersed in said liquids and means for withdrawing said second container from said first liquid into said second liquid at a uniform, predetermined, rate.

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