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# The Universal Multizone Crystallizator (UMC) furnace: an international cooperative agreement

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## ABSTRACT

The Universal Multizone Crystallizator (UMC) is a special apparatus for crystal growth under terrestrial and microgravity conditions. The use of twenty-five zones allows the UMC to be used for several normal freezing growth techniques. The thermal profile is electronically translated along the stationary sample by systematically reducing the power to the control zones. Elimination of mechanical translation devices increases the systems reliability while simultaneously reducing the size and weight. This paper addresses the UMC furnace design, sample cartridge and typical thermal profiles and corresponding power requirements necessary for the dynamic gradient freeze crystal growth technique. Results from physical vapor transport and traveling heater method crystal growth experiments are also discussed.

## 1  Introduction

The Universal Multizone Crystallizator (UMC) has been developed and patented by a team at the University of Miskolc for crystal growth of long duration space missions.[1] As a result of a joint agreement between scientists of the University of Miskolc through the Hungarian Space Office and the NASA Microgravity Science and Applications Division, a test project was contracted. On this basis a collaborative study of the UMC was undertaken at the Marshall Space Flight Center (NASA/MSFC), involving the participation of both US and Hungarian investigators. The overall objective of this project was to evaluate the UMC characteristics so as to assess the feasibility of utilizing the UMC for materials science experiments on the Space Shuttle, the Space Station and free-flying platforms.

Figure 1 shows the schematical layout of the UMC furnace and support equipment. The furnace consists of 25 independent resistance heated elements separated by fibrous ceramics. The outer furnace shell is held at a constant temperature by a water coolant loop. The furnace operates in a vacuum environment at $10^{-6}$ torr made by a CTI-Cryogenics micro-processor based cryopump.[2] The furnace is capable of processing ten samples by utilizing an automatic cartridge exchange mechanism controlled by an onboard computer. The cartridge
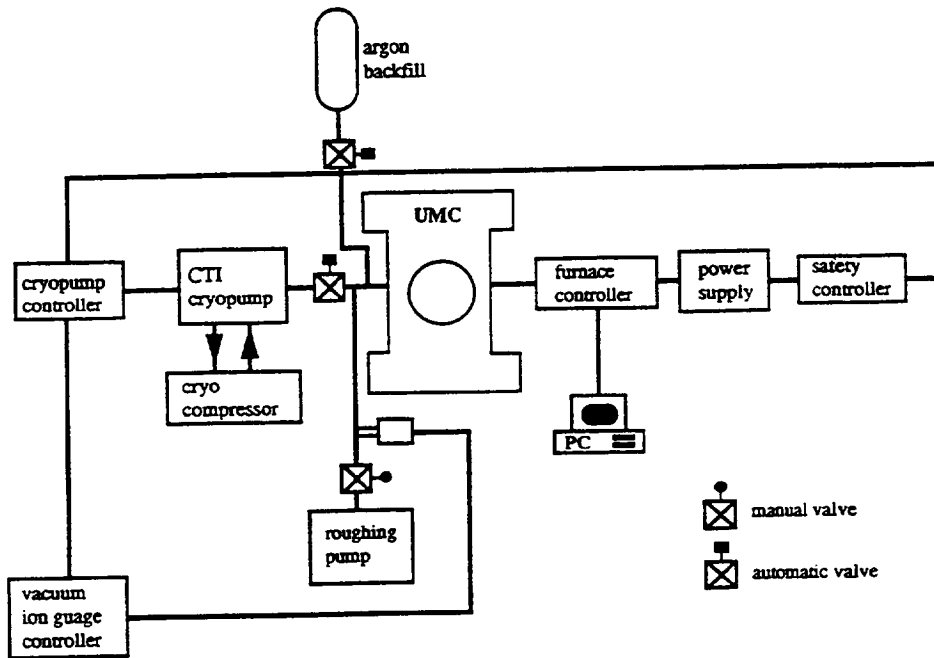
Figure 1: Schematic layout of UMC and support equipment.

depot can be removed from the furnace and returned to earth. Therefore, reducing mission costs by allowing the furnace to remain in space to receive further cartridges. A detailed description of the UMC is described elsewhere.[3] However, the main advantages and capabilities of the UMC are shown in table 1.

| Sample Dimension | 45 mm dia. x 250 mm max. |
|---|---|
| Temperature range | 1500 °C max. |
| Control accuracy | 0.1 °C |
| Translation rates | 0.02-10$\mu$m/sec |
| Furnace environment | vacuum |
| Typical growth cycle | 10 days |
| Cartridge depot | 10 cartridges |
| Sample exchange | automated |

Table 1: Operational specifications for the UMC.

# 2 Containment cartridge and experimental setup

In order to fulfill the different experiment requirements and taking into account the safety requirements of space flight missions, special cartridges have been developed for thermal testing and crystal growth.

The standard cartridge is shown in figure 2. It consists of a metallic tube with a wall thickness of 0.4 mm. A
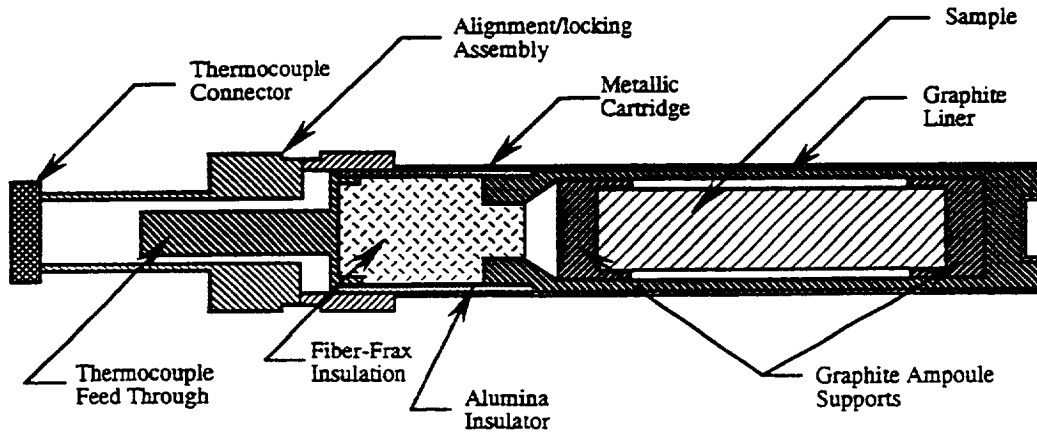


Figure 2: Schematic of standard processing cartridge with graphite liner.

graphite liner which can be instrumented with thermocouples is placed inside the metal tube. This liner is used to tailor the thermal profile in the furnace and can be exchanged with other materials as dictated by required thermal gradients. The sample to be processed is loaded into the graphite liner where graphite ampoule supports ensure that the growth axis is aligned parallel with the furnace bore. A maximum of 15 thermocouples can be used to monitor the crystal growth process. After the sample is loaded into the graphite liner, an alumina insulator section filled with fiber-frax is pinned to the bottom of the quartz liner. The insulator section limits the heat transfer to the thermocouple connection preventing erroneous measurements. Next, a stainless steel thermocouple feed through is pined to the alumina insulator. A keying mechanism is located on this feed through which allows one to determine the sample's circumferential location in the furnace bore. The cartridge is sealed by bolting a stainless steel alignment-locking assembly to the metallic cartridge. Lastly, thermocouples are connected to a temperature compensated connector located on the alignment-locking assembly.

For temperatures less than 1200 °C, a stainless steel cartridge is utilized. A tungsten cartridge is used for temperatures above 1200 °C. As the operating environment is vacuum, no cartridge coatings are required to prevent oxidation.

# 3 Typical thermal profile and power requirements

A typical thermal profile obtained during a dynamic gradient freeze experiment is shown in figure 3. For this
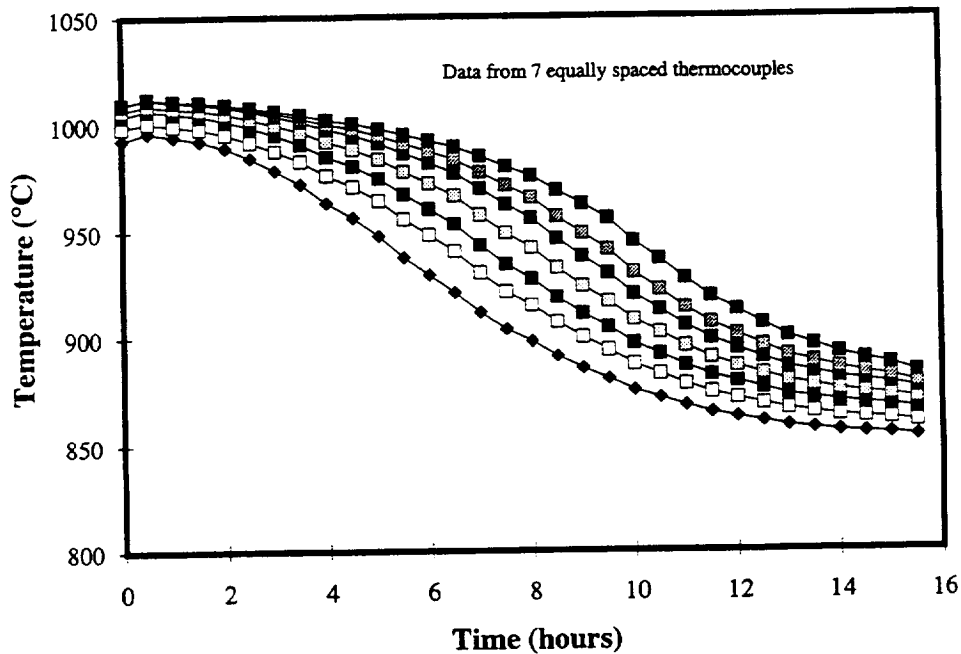


Figure 3: Thermal profile for dynamic gradient freeze experiment.

test the isothermal hot zone temperature was 1000 °C and the cold zone temperature was 850 °C. This produced a gradient in the furnace bore of 3 K/mm. As can be seen, the dynamic gradient freeze technique produces thermal profiles that are identical to the profiles produced by the Bridgman-Stockbarger[4,5] growth technique. The only difference is that in instead of mechanical translation of the thermal profile the dynamic gradient freeze technique systematically reduces the power to the individual heated zones. Thereby, directionally solidifying the crystal.

One of the main design constraints imposed on the UMC was to limit the power consumption. The maximum steady state power required during the above test was 450 W. Compared to other microgravity crystal growth systems,[10,11] the UMC requires approximately one-half the power for the same operating temperature. The power required for various set point temperatures is shown in figure 4. At an operating temperature of 1500 °C, the extrapolated power required is 850 W.
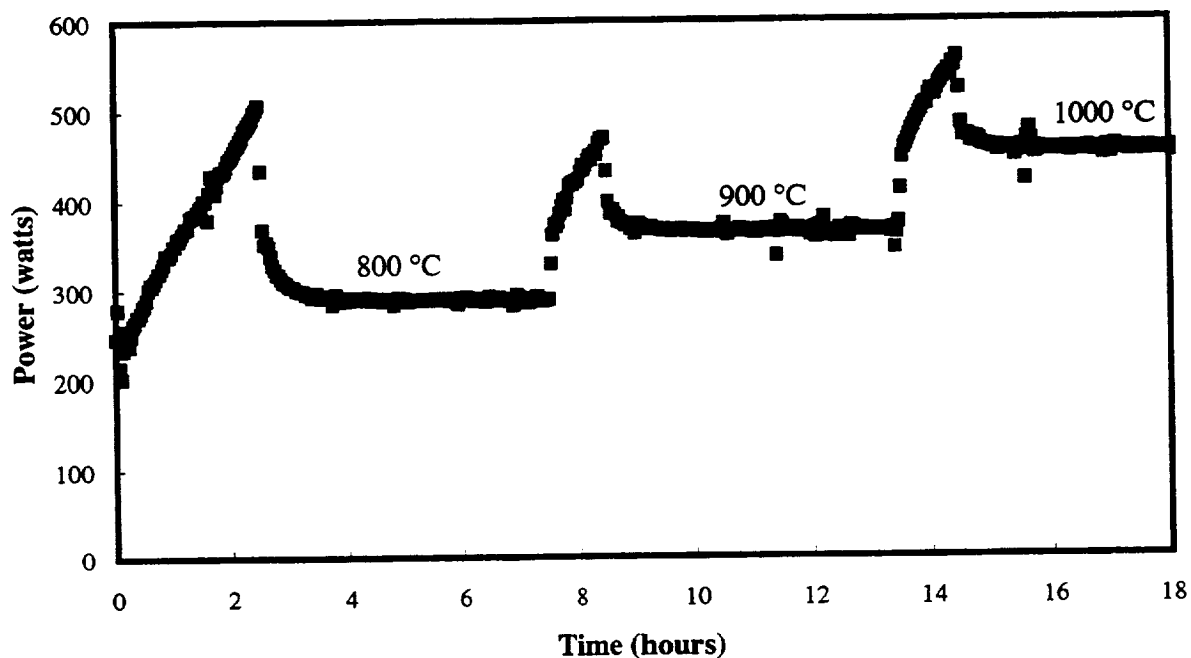
Figure 4: Power requirements for isothermal set point temperatures.

# 4 Crystal growth results

## 4.1 Growth of ZnTe by Physical Vapor Transport (PVT)

The PVT growth technique, in simple terms, involves the sublimation of a souce material. This source material is located at one end of the ampoule where it is heated above its sublimation point. It then transports to a cooler region of the ampoule where it crystallizes. The advantage of this technique is that the crystals tend to have a lower concentration of defect densities as compared to crystals grown from the melt. This is due to the fact that processing temperatures for the PVT process are significantly lower than the melting temperature.

To determine if the UMC could provide quality crystals utilizing the PVT crystal growth technique. A ZnTe PVT crystal growth experiment was performed. The starting material of ZnTe was distilled under vacuum from ZnTe powder which was synthesized from pure Zn (69 grade from Johnson Matthey) and Te (69 grade double zone refined from Johnson Matthey). The PVT growth procedures were developed by Su and colleagues and are given in detail elsewhere[12],[13] The growth was performed under a vertical stabilized condition, i.e., the hot zone on the top with respect to gravity. During the growth process the source was held at 1010 °C, a maximum of 1075 °C in the middle and a lower temperature of 925 °C in the growth zone. The thermal gradient at the interface measured was approximately 6 C/cm and the average growth rate was 4.3 mm/day. The grown crystal is shown in figure 5. All the starting source material was transported and a single crystal with a slightly concave (toward the crystal) growth interface was grown. The growth direction was determined to be 2° off (321) by x-ray Laue diffraction.
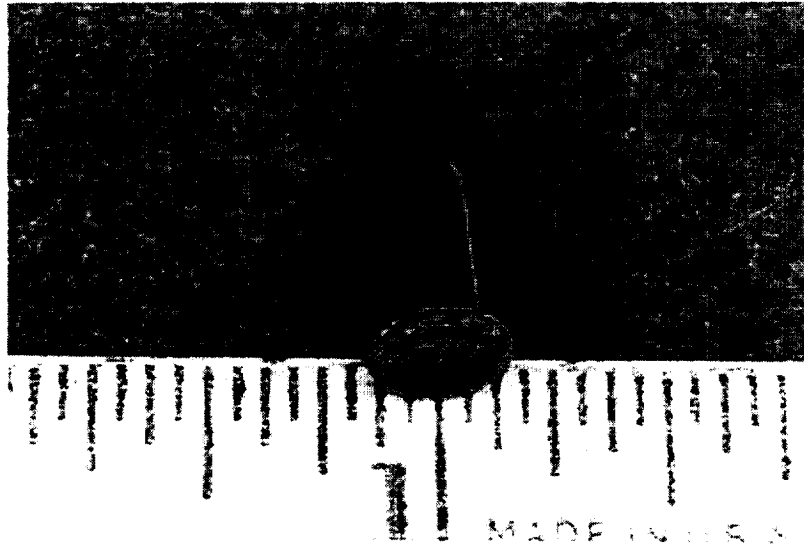
Figure 5: PVT growth of ZnTe.

## 4.2 Growth of CdTe by the Traveling Heater Method (THM)

Also called the traveling solvent zone method, the principle is to dissolve polycrystalline source material in a low melting point solvent zone, and then by translating the zone, deposit single crystal material on an oriented seed. As the temperatures involved are considerably lower than the melting point of the material, crystal quality is higher than from melt growth. In addition, purification of the starting material takes place as the zone passes through; the process is analogous to zone refining.

For this experiment the sample consisted of a 2 cm long, 8 mm diameter (111) oriented seed of CdTe, a 1.5 cm zone of tellurium containing 13 percent CdTe, and 10 cm of bulk CdTe. The diameter of the sample increased to 1 cm in the zone and the source material. The zone material was melted down under argon onto the seed. It contained 2.7795g Te and 0.3826g CdTe. This composition would be fully molten at 669 °C according to the phase diagram. The source material, in the form of a rod of CdTe, divided into four pieces , was then added above the tellurium zone and the entire sample sealed under high vacuum. In the UMC furnace, the zones were set up to establish a region 1.5 cm long with the temperature throughout higher than 669 °C. The procedure for translation of the temperature profile through the sample had to accommodate the finite widths of each zone. Therefore, a complex algorithm was developed to optimize the profile. The maximum set points of the zone was 725 °C, with the "background" temperature in the source and processed material kept a t 545 °C. This translated to a maximum estimated peak temperature of 704 °C in the solvent zone. The temperatures of the zones were programmed to translate the temperature profile through the sample at 0.064 mm/hr or 1.5 mm/day.

The growth was inadvertently terminated by a power failure after 21 days, or a translation of just under 33 mm. After removing the sample from the fused silica ampoule, the sample was as shown in figure 6. The surface of the sample was x-rayed with the back reflection Laue technique and it was confirmed that the sample was single
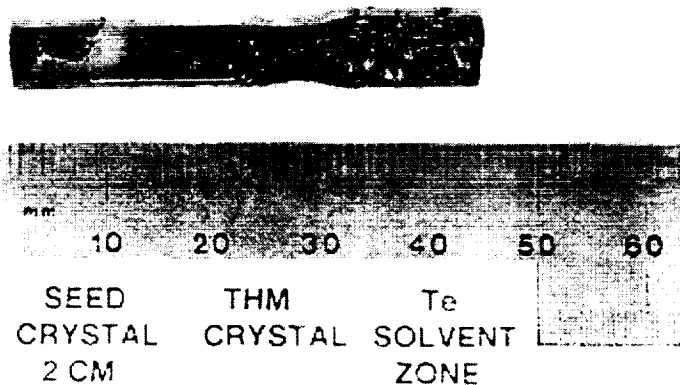
Figure 6: CdTe crystal grown by THM.

crystal and had the same orientation as the seed. It was then oriented and sliced on the (111) direction. The sample exhibited some voids at the boundary between the seed and the grown crystal. Structurally, the boundary was clearly defined by other parameters. Infrared microscopy showed an increase in tellurium inclusions in the THM crystal compared to the seed. The defect density was typical of CdTe and not very different from the seed crystal. Nevertheless, the UMC successfully translated the tellurium zone through the source material and was able to grow a single crystal of CdTe. Temporal modifications to the furnace zone temperatures could be refined to stabilize the size of the zone, the temperature gradient at solidification and hence improve the quality of the crystal. With optimized parameters, it is anticipated that a high quality crystal can be produced utilizing the UMC and the traveling heater method of crystal growth.

# 5 Summary

As part of an international cooperative agreement the UMC furnace capabilities were investigated at the Marshall Space Flight Center. The overall goal of the project was to determine if the UMC could be used to for materials science experiments. To this end, the UMC demonstrated that it was capable of producing single crystals with the PVT and THM crystal growth techniques. Additionally, it was shown that the UMC's power consumption is considerably less as compared to similar crystal growth furnaces.

# 6 Acknowledgments

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