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A STUDY OF SOLUTION CRYSTAL GROWTH IN LOW-g (2-IML-1)

Ravindra B. Lal
Alabama A&M University
Huntsville, AL USA

I. INTRODUCTION

During the International Microgravity Laboratory-1 (IML-1) mission it is planned to grow triglycine sulfate (TGS) crystals from aqueous solution using the modified Fluids Experiment System (FES). A special cooled sting technique[1] will be used for solution crystal growth. The objectives of the experiment are: (a) to grow crystals of TGS using the modified fluids experiment system, (b) to perform holographic interferometric tomography of the fluid field in three dimensions, (c) to study the fluid motion due to g-jitter by multiple exposure holography of tracer particles, and (d) to study the influence of g-jitter on the growth rate.

TGS cystals have technological interest as a pyroelectric infrared detector that can be used with no cooling devices. There are many applications for improved infrared detectors in military systems, astronomical telescopes, Earth observation cameras, and environmental analysis monitors. When grown to useful sizes on Earth, the crystals develop defects that limit their performance.

II. EXPERIMENT

During the IML-1 mission it is planned to have two experiment runs.

1. TGS-1 run (Isothermal) Sting and fluid temperature same and near saturation temperature. (010) oriented polyhedral seed (Figure 1).

2. TGS-2 run (Polynomial) The sting follows a predetermined polynomial. (001) oriented polyhedral seed (Figure 1).

A. Short Description of the Two Experiments

These experiments will be performed in the Fluids Experiment System (FES). The details of the FES are given elsewhere.[2] The FES is a windowed and instrumented crystal growth research cell designed to allow a variety of holographic diagnostics and schlieren viewing of the crystal and surrounding fluid as the crystal grows in space. Two types of images are recorded: schlieren images (transmitted on down-link as black and white video that reveals flow patterns and variations in fluid density) and holograms (recordings of three dimensional images that lead to quantitative determination of the concentration of the solution surrounding the crystal). TGS solution is nearly transparent, so it is possible to record holographic images through the fluid around a growing crystal. Using holographic interferometry, temperature and concentration gradients in the fluid and their motion can be monitored to determine how they are reduced in space and to determine how the crystal growth takes place in a low gravity environment. The FES has been slightly modified since the Spacelab 3 mission in 1985. The following modifications have been made to the FES. The knife edge has been realigned to avoid blackout at certain angles; holographic optical elements (HOE) have been added to the test cell windows for three-dimensional reconstruction of concentration gradients; a new software has been added; a viewing window has been added to view the test cell without opening the doors of the test cell enclosure; test cell bladder in the cap assembly has been reworked; a gold coated stainless steel pump has been replaced by a titanium pump; and the inside of the test cells has been coated with silicone RTV to reduce metallic contamination of TGS solutions.

The FES optical diagnostic system was modified for IML-1 after its last use in Spacelab 3 to incorporate holographic tomography (the ability to take optical data through the cell at multiple angles) as shown in Figure 2. Specially designed holographic optical elements (HOEs) have been added to the main test cell windows to provide a capability to illuminate and view the crystal at three different angles. The straight through view remains unchanged and two views at 23.5° are made possible by the HOEs. The schlieren system operation remains essentially the same while the hologram now records all three views on a single frame in a manner that allows them to be separated later back on Earth. The availability of the three views will lead to a more accurate quantitative understanding of the solution concentration and crystal growth.

Holograms and video data will be recorded during mechanical operations and critical phases of the crystal growth. Acceleration of the seed particles can be observed in real time on the video down link and will be determined accurately from the analysis of the holography data. During the experiment, the principal investigator and his team will monitor the down link video of the crystal and the growth solution and may instruct the crew to adjust some growth parameters.

In the first experiment a TGS crystal oriented in (010) direction will be grown for approximately 23 hours. The (010) face is the fastest growing face in TGS and is also the face required for detector fabrication. The growing face is uncut and unpolished and is about 1 cm x 1 cm in size. The solution of TGS will be seeded with buoyant particles of three different sizes (300 μm, 400 μm, and 600 μm). The main objective for this experiment run is to determine the effects of g-jitter and other g-variations on the fluid flow. Holograms will be taken at a programmed rate especially during any crew activities, and other Microgravity Vestibular Investigation (MVI) activities.

The second experiment run is basically a crystal growth run. The time allotted for this run is about 43 hours. No particles will be added to the TGS solution. The seed crystal will be (001) oriented polyhedral crystal which is uncut and unpolished on the growing side. This seed is about 1.5 cm x 1.5 cm in size. The experiment will follow a predetermined polynomial[3], but depending upon actual space conditions, the growth rate can be modified by changing the constants in the polynomial by crew interface. After the growth, the sting and the grown crystal will be removed from the test cell and will be stored in a specially designed container.

The payload crew will begin each run by placing a pre-stored sting with attached TGS seed crystal in a test cell filled with about 1.0 liter of TGS solution. The seed and the fluid will be separated by a cap assembly that will be retracted when the experiment begins. To dissolve all crystallites that will have formed in the solution at room temperature, the test cell will be preheated to a temperature of 70 °C on the optical bench, while keeping the sting temperature at 42 °C. The cell and the optical bench will then be cooled while the sting is heated to an appropriate temperature to bring the sting and the fluid to around 46 °C. At this temperature the cap will be retracted. A part of the seed will be dissolved to remove any surface imperfections and spurious nuclei. After that the experiment will follow a predetermined schedule. A typical time line profile is given in Figure 3. At the end of the run the cap will again be closed, the test cell removed from the optical bench, and the sting removed from the cell and stored in a specially designed container such that the crystal cools slowly to room temperature.

After the mission, the crystals will be returned to our laboratory for extensive investigations for their structural and electrical properties, including their capability for infrared detection. The space grown crystals will be compared with the laboratory grown crystals. Holograms taken during the mission will be reconstructed on ground for detailed optical analysis of fluid behavior. Data on the particle studies will be analyzed and correlated with "g" measurements on the shuttle.

On the IML-1 mission, accelerations that may cause convection and disturb the crystal growth will be carefully monitored. In addition to an internal FES accelerometer that will measure acceleration above 5 Hz, the Space Acceleration Monitoring System (SAMS) has two accelerometers located in the FES rack and one mounted in the Spacelab aisles. The special set of accelerometers are expected to provide information on low frequency (0 to 5 Hz) vibrations. Because the effects of g-jitter are believed to be extremely important in the crystal growth experiment we chose to incorporate optical techniques to monitor these effects directly.

III. UNDERLYING SCIENCE FOR THE GROWTH OF TGS CRYSTAL

A. Growth of TGS Crystals

The dielectric loss in TGS is due to the following: domain relaxation, microscopic solution and air inclusions incorporated during growth, and dislocations/strain centers which pin the domains. In space grown crystals we expect the growth to be mainly diffusion controlled and so we expect ordered deposition, fewer microscopic solution and air inclusions (solution is degassed before filling) and less strain centers and so domains will be more mobile. So we expect low dielectric loss (ε'') and improved figure of merit.

The ultimate use of these crystals is for IR detectors for 8 µm-14 µm range. The detectors will be fabricated at EDO/Barnes Engineering Division in Shelton, CT. All other electrical properties will be measured in our laboratory at Alabama A & M University. A section of both crystals will be analyzed by high resolution synchrotron X-radiation diffraction imaging at Brookhaven National Laboratory with the help of NIST.

A study of the effects of microgravity on TGS crystal growth in space by computer simulation is underway to help in the design of control parameters of spaceflight experiments.

A mathematical model of TGS crystal growth in space has been established including the field model, the interface model, and various boundary conditions.[4] The behaviors of the TGS aqueous solution follow the basic conservation laws of continuity, momentum, energy and concentration which are time dependent. The liquid-solid interface follows the growth kinetics and the mass balance. A finite volume code called COM-MIX has been employed and extensively modified to implement the simulation.

A series of two-dimensional cases are tested with different steady background g level ($10^{-1} \sim 10^{-4}\ g_0$) and different orientations (horizontal and vertical). Some cases of g-jitter are also simulated. Alternative boundary conditions are being tested. The results will be compared with the flight data.

B. Experiment with Seed Particles in the FES

Seed particles of three different monodisperse sizes will be added to the fluid in the FES. These will be observed on TV downlink in real time (only the largest particles are expected to show up clearly on the TV image). Their positions will be noted during each downlink and compared with later values to give us preliminary information on fluid and particle motion. The TV image will provide limited resolution and accuracy. Image processing will be done on line on the downlink image to help improve this. Holograms of the particle distribution will be made at preselected times during the experiments to later allow an accurate three-dimensional location on the particles as a function of time. Preliminary ground based results of particles image displacement velocity experiments were presented by Trolinger et al.[5]

1. Primary Science Objective

The primary science objective is to observe and quantify minute convection currents in the vicinity of the crystal and to correlate these with crystal growth processes. We will attempt to observe, isolate, and quantify the following components of fluid convection:

(a) Growth driven convection
(b) Convection due to g-jitter-random forces
(c) Convection due to other quasi-steady state acceleration forces - water dumps, etc.
(d) Convection due to residual microgravity - air drag, gravity gradient, etc.

The use of different particle sizes should allow us to make these measurements accurately and with high spatial resolution. The diffusion coefficient in g-jitter varies as the fourth power of radius, while terminal velocity varies as second power of radius.

We should be able to correlate the presence of convection with crystal growth anomalies and processes, and we should be able to correlate the presence of convection with other events. Namely, we should be able to more accurately characterize the space shuttle environment, how energy is coupled to an experiment, and its effect on crystal growth.

An accurate measurement of convection will be made in a regime never measured before. This will allow the validation of both theory and CFD codes.

2. Secondary Science Objectives

The dynamics of three ensembles of monodisperse particles in suspension will be observed in microgravity. This will provide important basic scientific information about inertial random walk, two phase flow, particle dynamics, and residual gravitational field in the Spacelab.

Testing the theory of intertial random walk - Inertial random walk, a novel type of diffusion, has been predicted by Regel et al.[6] to exist in microgravity, but has never been observed. It may have a significant effect on certain types of materials processing in space. It has been simulated (in one dimension) in ground experiments but only when measured in space will the theory truly be tested. Our particle experiments are almost ideal for testing the theory of inertial random walk. Requirements of particle sizes and types are fortunately similar for both the primary science objective and this objective, allowing us to pursue this objective with no added cost.

Testing two-phase flow and particle dynamics theory - Two phase flow has been studied very little in microgravity. The general equations of two phase flow are surprisingly untested because of the difficulty of testing many regimes. The microgravity environment will allow us to examine two phase flow in an extremely low Reynolds number regime where convection would normally dominate the results. The use of multiple particle sizes will allow us to test a variety of two phase flow effects.

Materials processing which involves free-floating particles depends heavily on particle dynamics and interaction. The statistics of particle diffusion and collision rates have been developed but have not been tested in microgravity. Although our selected particle number density is somewhat low, the study of particle dynamics and interaction should be enhanced by the tracking of all particles in three-dimensions in microgravity.

Residual gravitational field in Spacelab - The equations of motion of the particles lead us to the conclusion that we will be able to accurately measure the residual gravitational field on the Spacelab by tracking these suspended particles. The selection of the size and density range has increased the accuracy of the measurement. We should also be able to measure relatively low frequency changes in g. High frequency changes in g will be filtered out of our measurement. The measurement will be correlated with other recorded events in the IML-1 mission. This should provide a significantly improved characterization of the Spacelab environment.

C. Holographic Optical Elements (HOEs, holograms that emulate optical components)

First space application of HOEs - HOEs expand the FES optical system capability from a single angle of view to three angles of view without any significant weight or volume changes in the FES. The HOEs, thin materials which are attached to the FES windows, split the input light beam into three beams traversing the crystal surface at three angles. This is important to resolve any non-axially symmetric diffusion fronts. The HOE on the output window collects the three beams and bends them back to allow their collection in the same recording camera. To achieve the same thing with conventional optics would have required a major redesign and configuration change in the FES and would have added considerable weight and volume. The concept is likely to find application in other optical diagnostics requirements for space experiments where weight and volume are critical parameters, and where a wider angle of view is needed for viewing.

ACKNOWLEDGEMENTS

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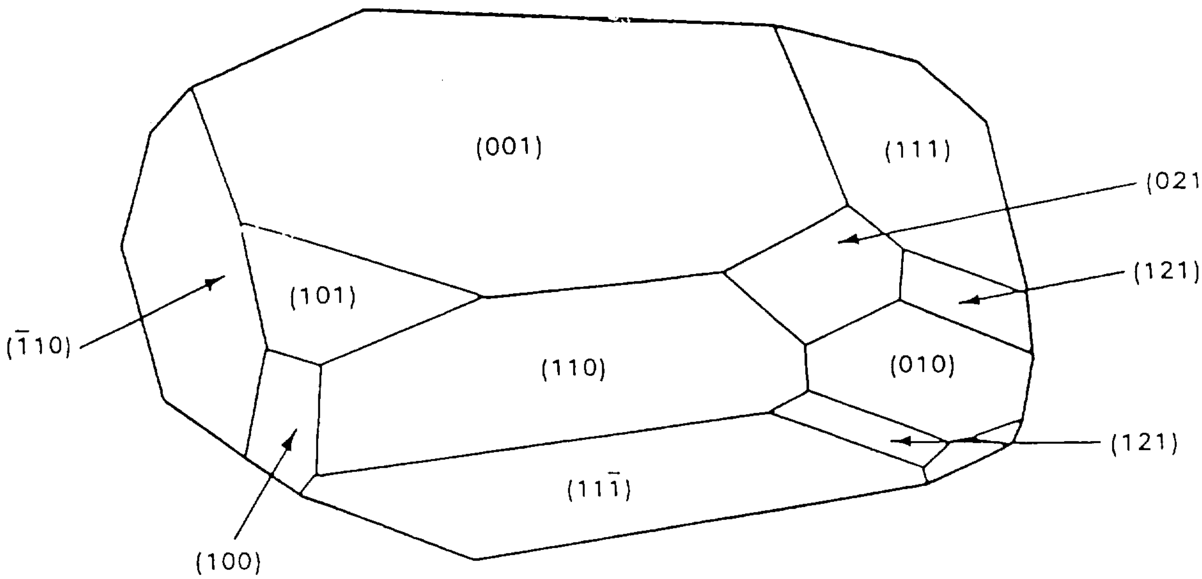
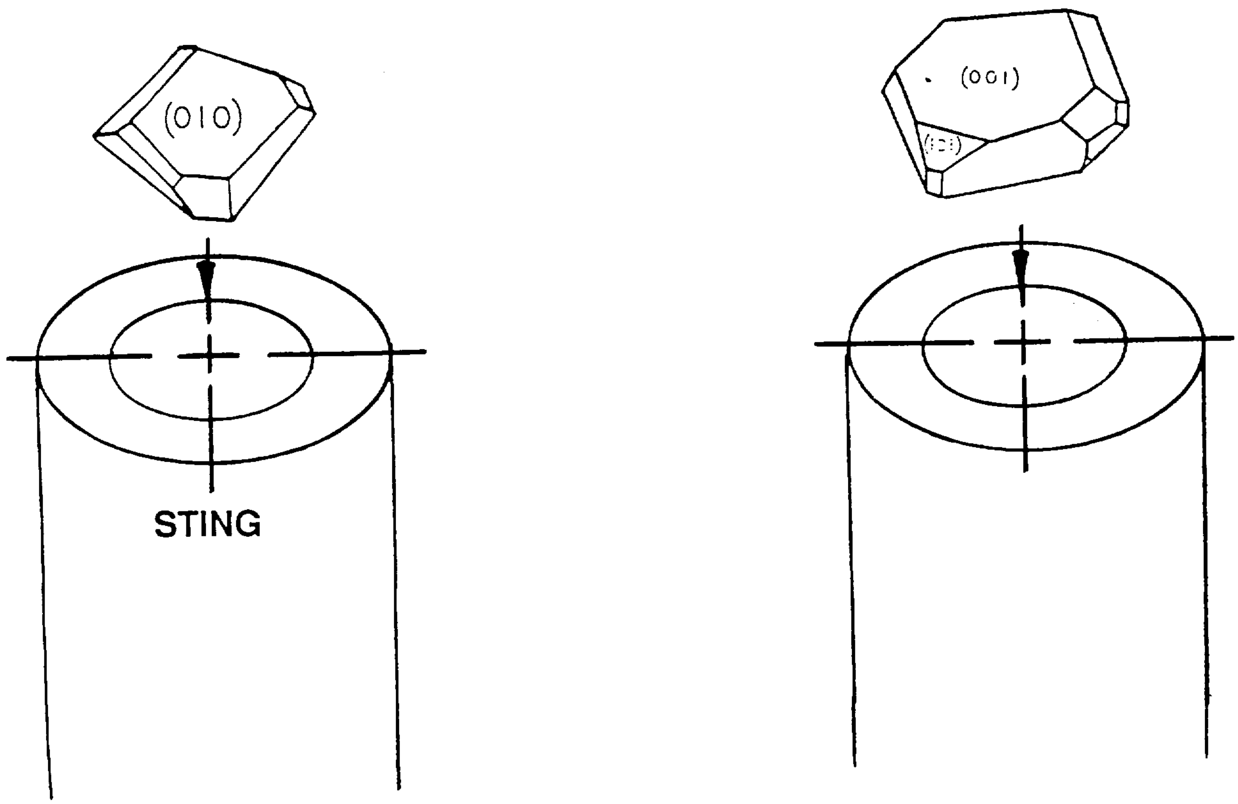


CRYSTALLOGRAPHIC FACES OF TGS CRYSTAL



TGS-1

TGS-2

Figure 1. Triglycine Sulfate Growth Runs on the IML-1 Mission.

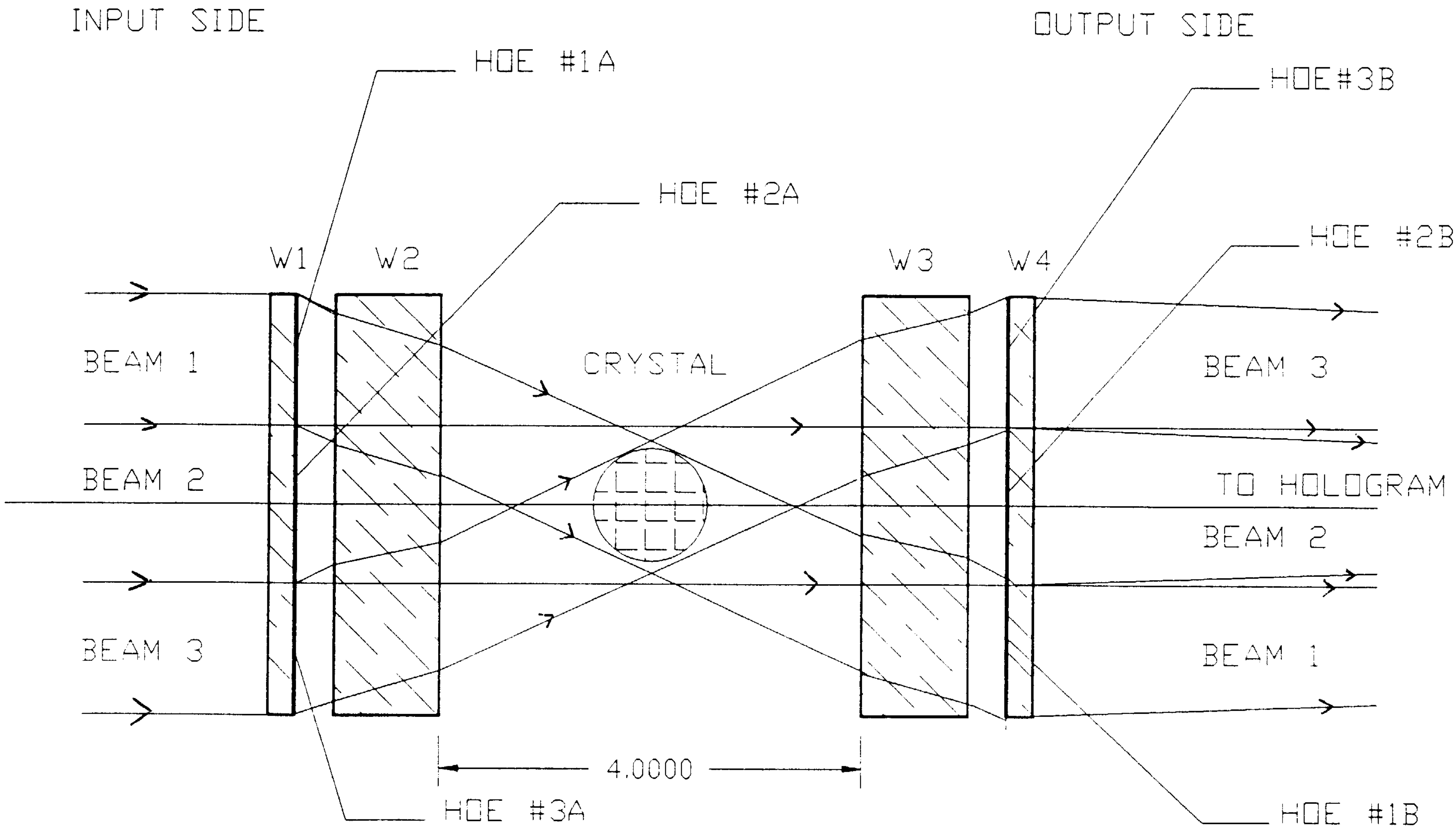


Figure 2. FES Optical Diagnostic System.

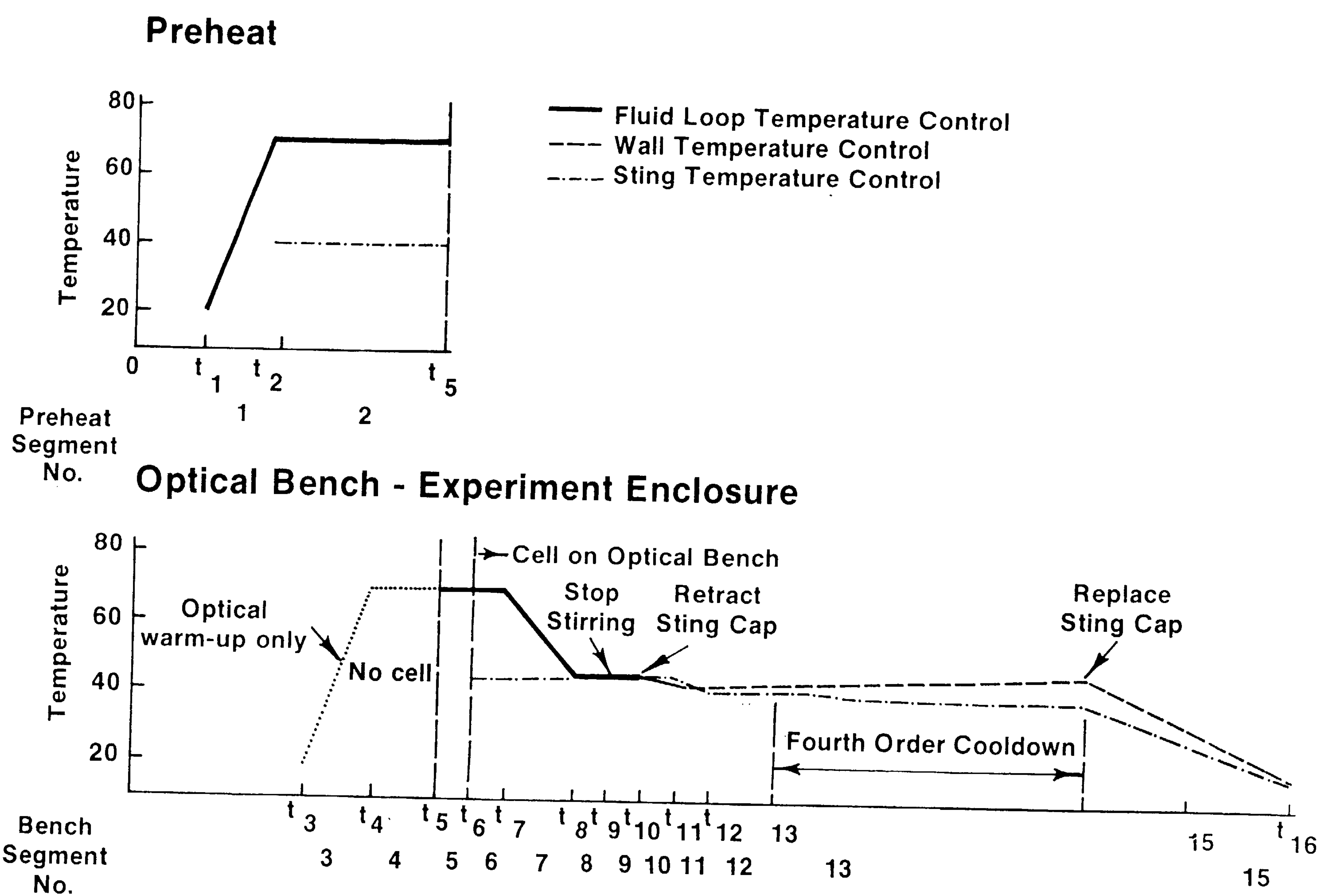


Figure 3. FES Timeline.