Void Management in MEPHISTO and Other Space Experiments

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The second flight of NASA's Shuttle Flight experiment program known as MEPHISTO suffered from a void in the liquid portion of the sample, even though a piston arrangement was in place to keep the ampoule filled. In preparations for the next flight of the MEPHISTO furnace an animated computer program, called MEPHISTO Volume Visualizer (MVV), was written to help avoid the formation of unwanted voids. A piston system on MEPHISTO has the ability to move approximately 5 mm in compression, to accommodate expansion of the solid during heating; then from the completely compressed position, the piston can move up to 25 mm in towards the sample, effectively making the ampoule smaller and hopefully eliminating any voids. Due to the nature of the piston design and ampoule and sample arrangement, the piston has gotten stuck during normal directional solidification; this creates the risk of a void. To eliminate such a void, the liquid in the hot zones of the furnace can be heated, thereby expanding the liquid and consuming any void. The problem with this approach is that if the liquid is heated too much an overpressure could result, breaking the ampoule and ending the experiment catastrophically. The MVV has been found to be a useful tool in the assessment of the risks associated with the formation of a void and the additional heating of the liquid in the hot zone of this Bridgman type furnace. The MVV software will be discussed and copies available; it is written in the Delphi 2 programming language and runs under Windows 95 and NT. The strategies used in other flight experiments, such as the Isothermal Dendritic Growth Experiment, will also be presented.

Introduction

Many fluids and materials microgravity experiments have experienced failures due to the presence of voids. Voids, in addition to displacing the desired phase, also cause problems with heat transfer, convection caused by gradients in surface tension (Marangoni convection) and blocking clear views to areas of interest. On SkyLab, undesired voids or bubbles were present in four materials experiments.[1)] Voids are believed to have caused significant Marangoni convection in a recent lead-tin-telluride crystal growth experiment,[2)] caused complete mixing rather than the diffusion controlled state desired in a selenium doped GaAs experiment,[3)] and loss of the high thermal gradients required in a Al-In solidification experiment.[4)] The experiment associated with the second flight of the directional solidification furnace known as MEPHISTO-2 was hampered by voids which caused a break in the continuity of the cylindrical Bi-Sn sample.[5)] This void caused a loss of the Seebeck signal[6,7)] - which was the primary measurement of the experiment.

Many experiments, such as the Isothermal Dendrite Growth Experiment and MEPHISTO-4, were successful with their plans and efforts to avoid and accommodate voids. This paper will discuss some of the successful technologies and strategies used to eliminate void space and, for example, accommodate solidification shrinkage during space experiments, while concentrating on the techniques and tools used on the MEPHISTO furnace system. A Delphi 2 computer program is presented which estimates and predicts voids and overpressure in cylindrical samples.

Procedures for void accommodation

In general, we will consider two basic types of voids: 1) those that are vacuous - containing only the vapor pressure of the surrounding liquid, such as those due to solidification shrinkage (surrounded by liquid) or left behind after a good (10-4 torr) vacuum is drawn on a container before sealing; and 2) those with a gas that can not be absorbed into the surrounding liquid. Usually some void space is required either due to engineering difficulties associated with containment and sealing the container or to accommodate expansion of the liquid during heating.

One of the simplest ways to eliminate free surfaces and voids is to contain the sample in an evacuated container equipped with a spring loaded plunger. Figure 1 shows a design proposed for Shuttle flight experiments.[8,9)] The outer container is welded stainless steel with an evacuation tube and a thermocouple with a stainless steel sheath brazed in with a support button. The inner container is boron nitride with a close fitting (less than 0.0025 cm total clearance) plunger driven by a tungsten wire spring. The plunger is: hollow so the spring fits into

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it and does not buckle; and long so it does not get tilted or stuck. High quality surface finishes (64 rms) on the mating surfaces of the boron nitride help insure smooth operation. If considering solidification with heat losses nearly the same from all walls - an unavoidable shrinkage cavity may form near the center of the sample - unless the sample is compressed in the mushy state to collapsed shrinkage voids. If considering approximately directional solidification care should be taken to arrange the plunger at the hot end, so the plunger and spring do not have to push and move the solid to accommodate solidification shrinkage. In Figure 1 the hot end of the furnace was on top, with cooling from below, so the plunger is always at the hot end of the ampoule. Boron nitride is an excellent crucible material, however, it is also expensive. An alternative material is a machinable ceramic such as MACOR. One of the most importance aspects for success here is achieving a good vacuum while sealing the container. Even a very weak spring and plunger will collapse a void if the spring/plunger has enough movement available, the sample is all liquid, and the void contains only the vapor of the surrounding liquid. If there is a foreign gas in the void - resulting from a poor vacuum, or internal degassing of crucible materials for example, even a very forceful plunger can not be expected to eliminate it.

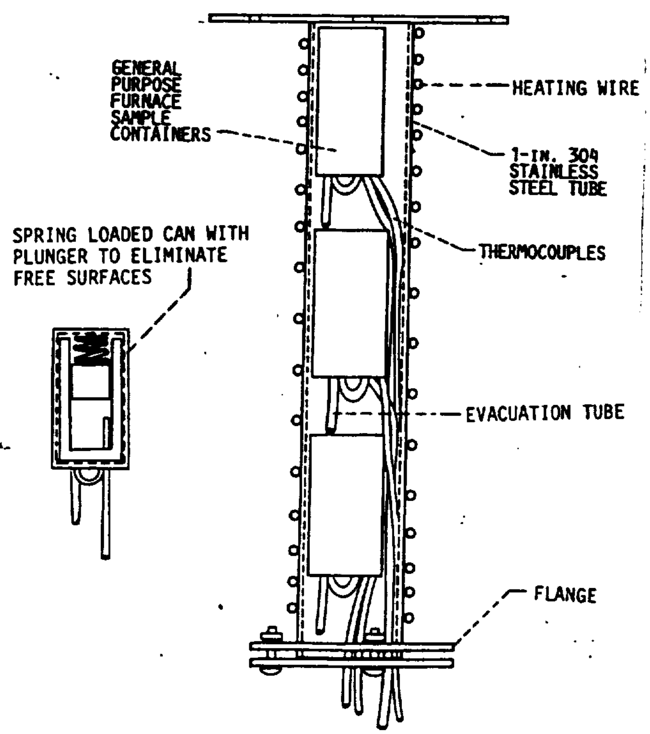

Figure 1 Furnace and sample container schematic (from Ref. 8). Stainless steel sample container cross-section shows thermocouple in the sample, boron nitride crucible and plunger, and tungsten wire spring. The plunger helps to eliminate voids and liquid/vapor free surfaces and thus Marangoni flows.

Figure 2 is a schematic of the PbSnTe samples processed in space in the Advanced Automated Directional Solidification Furnace (AADSF).[2] The samples are contained in fused silica. No need for eliminating free surfaces was anticipated. In addition to the voids expected at the ends due to partial filling of the ampoule, it is believed that there was some sticking of the liquid to the ampoule ends - thereby stretching the liquid out and exacerbating the deleterious effects of the void space present in the ampoule. Longitudinal composition analysis of one of the samples showed the concentration profile along the length of the sample to be characteristic of solidification into a fully mixed liquid.[2] A previous space experiment in this series also suffered from a high degree of mixing.[10] It is likely the mixing in these experiments was a result of the presence of void space which enabled Marangoni flow and sloshing of the liquid in the ampoule. Modeling and scaling analyses have estimate only moderate mixing even with gravity at 10^{-5} g_o directed 90 degrees to the ampoule axis;[11,12] thus it is unlikely the mixing in these experiments could have been a result of any residual steady accelerations in flight.

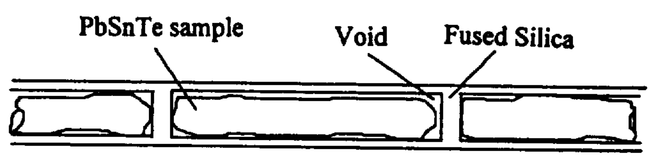

Figure 2 Schematic of a PbSnTe sample and fused silica ampoule flown in space in the Advanced Automated Directional Solidification Furnace (USMP-3, Feb. 1996). No methods were used to completely fill the ampoule, void space was allowed and planned. Compositional analysis showed the liquid to be completely mixed during solidification where diffusion conditions were the goal and expected. Mixing was likely the result of the void space which enabled Marangoni flow and/or sloshing of the sample in the ampoule due to acceleration disturbances know as g-jitter.

Void space and volume changes in the Isothermal Dendrite Growth Experiment (IDGE) flown on the Space Shuttle flight STS-62, March 1994, were accommodated through use of a stainless steel bellows,[13] as shown schematically in Figure 3; this worked, with the experiment performing flawlessly. Succinonitrile (SCN) was used in this undercooling and solidification experiment.

A dendrite growth and observation chamber used in the IDGE Shuttle flight experiment on STS-87, Nov. 1997 (USMP-4) is shown in Figure 4; this experiment used Pivalic acid (PVA). Steel chambers could not be used with PVA. Some void space was required in this fused silica growth chamber to prevent rupture upon heating of the liquid. The location of the void is manipulated using the compensator assembly which includes a piston made of an iron rod encapsulated in silica.[14] The precision (tight) fitting piston is moved using a set of solenoids. When the piston is moved towards the sample the void containing PVA vapor is encouraged to form (with a characteristic *crack*) on the low pressure side of the piston - thereby removing it from the observation chamber were the dendrite growth is taking place.

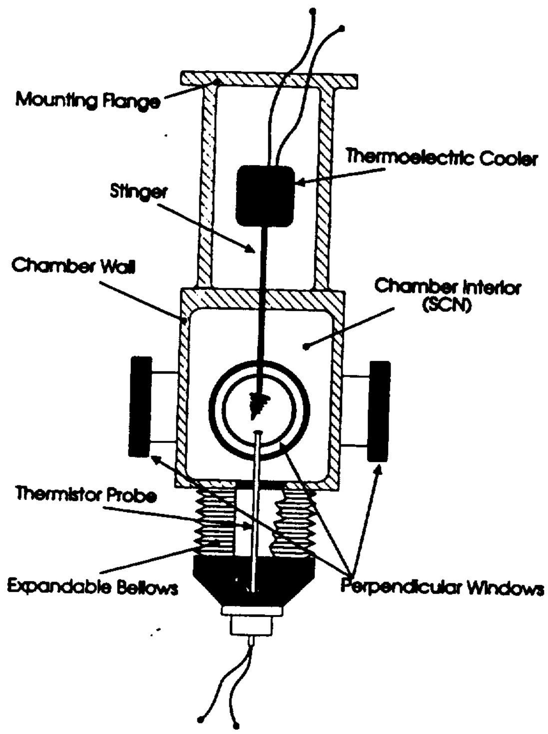

Figure 3 Schematic of the growth chamber used in IDGE on the USMP-2 mission, STS-62. Void space is eliminated by filling the chamber with solid under vacuum; then when the sample melts and expands any remaining voids are eliminated and further expansion is accommodated by expanding the bellows.[13]

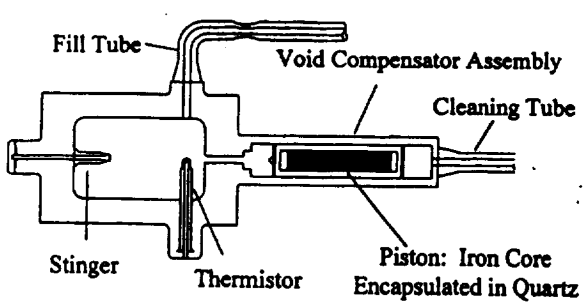

Figure 4 Schematic of the IDGE growth chamber and volume compensator assembly; stinger and thermistor are in the test fluid, voids are moved to the far side of the iron cored piston by moving the piston towards the chamber using a set of solenoids.

Void management on MEPHISTO

Bridgman type directional solidification experiments were done using the French built MEPHISTO furnace on all four of NASA's USMP missions. The second flight of the MEPHISTO furnace (MEPHISTO-2) suffered from a void in the liquid portion of the sample, even though a piston arrangement was in place to keep the ampoule filled. There were strict specifications from the French on the size (importantly, the diameter) of the Bi-Sn sample, so that it could be safely input into the fused silica ampoule. The cylindrical samples were approximately 900 mm long with a diameter of 5.8 mm. The fused silica ampoules were 6 mm internal diameter. Upon melting, the liquid needed to fill the whole 6 mm dia. ampoule; however, in addition to needing to fill the void present due to the difference between the ampoule and sample diameters, void space is also created during melting because the Bi sample contracts upon melting. The length compensator arrangement, shown in Figure 5, on the sample end was designed to push additional sample into the hot zone until all void space was consumed. For MEPHISTO-2 there was not enough available movement in the compensator to fill the ampoule with liquid. As a result, a break in the liquid formed which resulted in an open circuit and the loss of the Seebeck signal.[6,7] There was also about a 200 mbar argon pressure in the gap between the ampoule and sample. This added the possibility that the void in the liquid could contain argon gas - making elimination of it (the argon bubble) nearly impossible. For the fourth flight of the MEPHISTO furnace the initial diameter of the solid sample was increased to 5.88 mm; this decreased the volume compensation need upon melting to a value within the capabilities of the compensator.

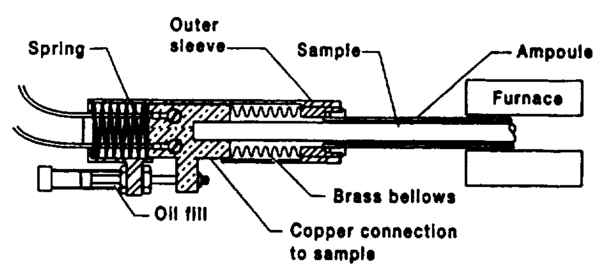

Figure 5 Schematic of the length compensation devise used on the MEPHISTO furnace. The spring pushes on a precision fit piston and rod, which move slowly due to the oil slipping around the tight fitting piston. The rod pushes the copper connector, collapsing the bellows and moving the sample further into the ampoule. The spring applies approximately 45 N to the piston. The piston moves approximately 5 mm in compression, to accommodate expansion of the initial solid during heating; then, from the completely compressed position, the piston can move up to about 25 mm.

During solidification, solid of 6 mm diameter is being formed. In order for the compensator to push the solid into the liquid it must overcome the friction forces between the 6 mm solid sample just solidified and the 6 mm ampoule. This creates the risk of the 6 mm solid sample getting stuck, immobilizing the compensator and allowing a void to form. To eliminate such a void, the liquid in the hot zones of the furnace can be heated, thereby expanding the liquid and consuming any void. The problem with this approach is that if the liquid is heated too much an overpressure could result, breaking the ampoule and ending the experiment catastrophically. An animated computer program, called MEPHISTO Volume Visualizer (MVV), was written to help avoid the formation of unwanted voids and help with the assessment of the risks associated with the formation of a void and the additional heating of the liquid in the hot zone of this Bridgman type furnace; it is written in the Delphi 2 programming language and runs under Windows 95 and NT.

The calculations done by the MVV are based on a table containing a series of steps input by the user. Each step includes the position of the moving furnace, the temperatures of the moving and fixed furnaces and an optional step name. The simulation progresses from step to step, calculating the position of any solid-liquid interfaces, the position of the piston, and the magnitude of any void or overpressure. The program provides facilities to create and maintain the step table as well as run the simulation. The user can step through the simulation under manual control or allow the program to automatically step through it using a specified delay between steps.

The results of the calculations are displayed graphically throughout the operation of the program and can also be printed. While interface positions are stored cumulatively, all other parameters are recalculated from steady-state conditions for every step.

MVV was intended to be familiar to those who are accustomed to programs running under Microsoft Windows. Its File menu provides the ability to clear the step table (New) allowing the user to create a new step file in the editor, open an existing step file (Open), save a step file(Save and Save As), restart the simulation and to print the results of the simulation. The Edit menu opens an editor that allows the user to create and edit the series of experiment steps in an interactive manner.

MVV was designed to be flexible in the definition of the sample and furnace parameters. Many of the material or physical parameters of the experiment can be modified to account for differences in experimental details from one use to the next. The Sample Setup menu allows the user to specify parameters for ampoule dimensions, constraints imposed by the length compensator, and the physical constants used for calculating sample density. If the simple density correction scheme (linear with temperature) used in MVV is inadequate for a particular material, the calculations for liquid and solid densities can be modified in the source code. They are written as separate functions residing in the MephData module. They require the temperature as a formal argument and return the density at that temperature. This arrangement allows them to be changed with minimal impact on the rest of the program.

Finally, the Furnace Definition menu pick brings up a window where a number of the physical dimensions of the furnace can be specified. These allow a number of furnaces of the same basic design to be tried, as parameters like the overall length of the furnace, the length of the hot zones, etc. can be easily modified. The real constraint on this configurability is that the "new" furnace design has to be defined in terms of the existing MEPHISTO hardware. New furnace segments cannot be added using the Furnace Definition window, for instance.

In order to modify the code to perform outside the existing MEPHISTO configuration one must know how the code is set up for MEPHISTO and thereby get a better understanding of what in the code needs to be modified to make it fit the particular application. The MEPHISTO furnace has two furnace assemblies, each with a hot zone, gradient zone, and chill zone. One of the furnace assemblies can move thereby inducing solidification or melting and the other does not move and is maintained as a reference for the Seebeck temperature measurement at the s/l interface. This requires that the volume, at constant mass, of six regions be determined (in addition to other things such as s/l interface location). These regions are, on the moving furnace – the chill, gradient, and hot zones; on the fixed furnace – hot and gradient zones; and last, the zone between the two hot zones. This is shown schematically in Figure 6 with the variable names as they are in the code. The length of the moving furnace chill zone and the length between the hot zones varies with furnace position. If, for example, the code was to be modified to solve a Bridgman type problem with single chill, gradient, and hot zones, with the lengths of the chill and hot zones varying with furnace position, there would be no code changes to ChillLength, LenChillGradZ would be changed to the new length of the gradient zone, and LenMovingIsothermalZ + LenFixedIsothermalZ + LenFixedChillZ would be made to equal the new length of the furnace hot zone and LengthBetweenFurn would equal the maximum throw or movement of the furnace; the temperature of the moving furnace chill would be set to the new chill value, and the temperature of the fixed furnace and the fixed chill would be set to the new hot zone temperature. In this example (as in MEPHISTO) at a furnace position of zero the minimum of the gradient zone is at the bottom, or end of the domain.

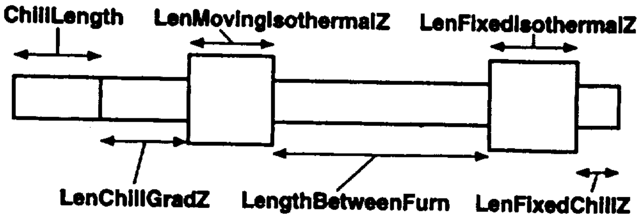

Figure 6 Schematic of the sample domains with variable names as they are in the code MVV.

In addition to the above procedures for void accommodation, there are some other alternatives to compensate for solidification shrinkage and sample expansion; including, for example, applying a smart material or intelligent structure to replace the spring and plunger of Fig. 1. The desirable smart mechanism should contract during heating, thereby allowing more volume for the expanding sample, and expand its volume upon sample cooling to make up for solidification shrinkage and sample contractions. If a void space is inevitable due to the sealing process or some other technical requirement, a suitable inert and viscous glass or fluxing agent can be used between the void and sample to eliminate the free surface at the sample and consequently reduce undesirable Marangoni convection. This can be taken further so that the whole sample is contained in a very compliant glassy container; this is done in undercooling experiments were the sample is surrounded by glass. The glass also works as a fluxing agent dissolving and otherwise rendering ineffective nucleation sites. Most of the sample processing takes place above the glass transition temperature, thus the glass is compliant, allowing the sample to expand and contract. Yet due to the high viscosity of the glass, a near no slip boundary condition is expected at the glass – sample interface.

Conclusions

Many fluids and materials experiments have been compromised due to the presence of voids and have shown the need to eliminate voids to prevent displacement of the desired phase, Marangoni flow, disturbance of the expected heat transfer, and visual obstructions. Two basic designs were presented here to accommodate volume changes, one autonomous consisting of a piston and spring, and a second with active control using an iron cored piston and solenoid. Both have been built, tested and used successfully. The container and sample volumes of a Space flight experiment should be determined and controlled if need be throughout processing so the deleterious effects of voids can be avoided. A computer program, MEPHISTO Volume Visualizer (MVV), for this purpose for the MEPHISTO-4 flight experiment was presented and has been found to be a useful tool in the assessment of volume compensation for MEPHISTO. The MVV program can be modified and used for other cases of cylindrical geometry.

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