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Initial Study of Void Formation During Aluminum Solidification in Reduced Gravity

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Void formation due to volumetric shrinkage during aluminum solidification was observed in real time using a radiographic viewing system in normal and reduced gravity. An end-chill directional solidification furnace with water quench was developed to solidify aluminum samples during the approximately 16 s of reduced gravity ($\pm 0.02g$) achieved by flying an aircraft through a parabolic trajectory. The aluminum was contained in a vacuum sealed, rectangular, pyrolytic boron nitride crucible. An ullage space was present during each test. Void formation was recorded for two cases: 1) a nonwetting system, and 2) a wetting system where wetting occurs between the aluminum and the crucible lid. The void formation in the nonwetting case is similar in normal and reduced gravity, with a single vapor cavity forming at the top of the crucible. In the wetting case in reduced gravity, surface tension causes two voids to form in the top corners of the crucible, but in normal gravity only one large void forms across the top.

Nomenclature

g = gravitational constant
k = thermal conductivity
r = characteristic length
ρ = density
σ = surface tension

Subscripts

l = liquid
s = solid
w = wall

Introduction

THE general problem of void formation in reduced gravity applies to thermal energy storage for space power and materials processing applications such as casting and crystal growth. Several planned space power systems require melting and solidification of a thermal energy storage material that undergoes a significant volume decrease when cooling from liquid to solid, thus causing voids to form. Specific examples of such phase change materials (PCM) are LiF-20CaF_2,[1] proposed for the solar dynamic power module of Space Station Freedom, and lithium,[2] proposed for a space nuclear power system. LiF-20CaF_2 has a 20% volume decrease when solidified and lithium contracts 25% when solidified. Also, from liquid/vapor studies in reduced gravity,[3,4] the ullage in the molten state (which eventually becomes a void when the PCM solidifies) may be in a variety of locations. The impact of voids in the PCM, specifically their uncontrolled or unknown location due to reduced gravity effects, raises two major concerns. First, if voids form in the solidified PCM adjacent to the heat input area, hot spots may result on the container walls. Second, if voids form in the solidified PCM away from the heat input area, there are no neighboring voids for the melting PCM to expand into; this will result in undesirable container stresses.

Two reduced-gravity experiments have investigated bubble motion and void formation from dissolved gases and material shrinkage during melting and solidification. A multiphase dispersion experiment[5] was conducted on the Space Shuttle by directionally solidifying cesium chloride (CsCl) that contained lead (Pb) particles and air enclosures. Another solidification experiment was conducted on a sounding rocket[6] using carbon tetrabromide (CBr_4) saturated with Ar, H_2, and N_2 gases. The transparency of the liquid phase in both experiments allowed photographs of the solidification process; this revealed nucleation and growth of bubbles which became voids. Gases were rejected during solidification and formed bubbles at the growth interface. In normal gravity the bubbles often departed the solid/liquid interface, leaving the specimen generally free of large internal voids. In reduced gravity a greater void density was observed in the solidified samples. In both of these experiments, bubble formation was due in part to the gas originally dissolved in the sample, in part to the volumetric shrinkage of the melt during solidification, and in part by the initial ullage volume.

Two future Space Shuttle experiments are being built to study void behavior for specific space systems. The Thermal Energy Storage (TES) Technology Experiment uses lithium fluoride (LiF), a high temperature, high heat-of-fusion PCM that will undergo four freeze-thaw cycles in low Earth orbit. LiF has a 29% volume reduction upon solidification and is a representative PCM for space solar dynamic power systems.[2] The sodium-sulfur (Na-S) Battery Flight Experiment will study two areas: 1) the wicking functions for the transport of electrode materials to the electrolyte interface; and 2) the cell's ability to accommodate freeze-thaw cycles in space since sodium has a 5% volume decrease and sulfur has a 12% volume decrease upon solidification.[2] Since neither the TES nor Na-S experiments have real-time viewing systems, the voids will be inspected during the post flight analysis.

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The purpose of the present experiment is to investigate the phenomena of void formation, growth, and motion due to volumetric shrinkage and liquid/vapor reorientation of the ullage during solidification of aluminum in reduced gravity. Reduced gravity is achieved by flying an aircraft through a parabolic trajectory. Aluminum was selected for this experiment due to its high thermal diffusivity and large volume change during solidification (7%) as compared to other conductive materials. This experiment differs from prior reduced-gravity work because no dissolved gas is present in the molten aluminum, thus avoiding the difficulty of distinguishing evolved gas from shrinkage caused voids.[7] Also, this experiment includes real-time observation of the solidification process, a feature not provided for on experiments being built for near-term Space Shuttle missions.

Experimental Hardware, Procedure, and Test Conditions

An end-chill directional solidification furnace with water quench has been developed and tested to solidify aluminum samples during approximately 16 s of reduced gravity. Solidification is recorded using a real-time x-ray unit and video system. The liquid/vapor and solid/liquid interfaces can be seen on the recorded image.

The reduced-gravity experimental data was obtained through the use of the NASA Lewis Research Center Learjet aircraft. The aircraft, with the experiment onboard, was flown through a parabolic trajectory as shown in Fig. 1. Acceleration was measured on the aircraft in the X, Y, and Z directions using three sensor heads and the output signals were recorded on a chart recorder. During each trajectory the acceleration range (gravity level) was maintained to within $\pm 0.02g$ in all three directions.[8]

Real-Time Radiographic Viewing System

The real-time radiographic viewing system includes the x-ray and image intensifier units, a high-resolution charged coupled device (CCD) camera, and a Super-VHS recorder, as illustrated in Fig. 2. The x-ray unit generates x-rays by accelerating electrons from a filament onto an anode using a voltage up to 80 kV at 2 ma; the excitation of the anode by the electron beam generates the x-radiation.[9] The focal spot size on the anode is 0.05 mm. In this study, an x-ray intensity of 40 kV was used to penetrate the crucible and 0.6-cm-thick aluminum sample. Although the aluminum sample should be as close to the image intensifier as possible, thermal limitations resulted in placing the furnace about midway between the x-ray unit and the intensifier.

The image intensifier serves as a wavelength converter, accepting x-rays and producing visible light. Within the image intensifier the x-rays strike a photoelectric coated screen which emits electrons when struck by x-ray photons. The electrons are accelerated and focused by electrostatic lenses onto a second, smaller but brighter fluoroscopic screen. This screen's output light intensity is suitable for pickup by a video system.[9] It is optically coupled to a high-performance video camera that converts the visible images to an electronic signal. The black and white CCD camera has horizontal and vertical resolution of 570 and 485 lines, respectively. The video images are stored on the S-VHS recorder or viewed directly from the monitor. Knowing the crucible dimensions, the smallest recognizable void measured from a radiographic image is approximately 1 mm.

Furnace Quench System

The furnace quench system includes a three-zone furnace, crucible, aluminum sample and water quench. The furnace, shown in Fig. 3, uses nickel-chromium alloy resistive heating elements with a maximum power of 200 W/zone and is capable of heating samples to 725°C. One thermocouple per zone is required for control purposes and to establish initial temperature conditions. Temperature data from a maximum of seven thermocouples can be recorded. Interchangeable crucibles having a volume up to $2.5 \times 2.5 \times 1.25$ cm for the samples are mounted in a fixed position. The samples are directionally solidified by spray quenching the bottom of the crucible with water. The water is collected in a reservoir tank and pumped back to the crucible at a flow rate of 8.5 l/min. The typical solidification rate of an aluminum sample is 0.133 cm/s.

Furnace Control System

The computer, data acquisition and control system (DACS), and power control unit (PCU) are used to retrieve temperature data and to provide regulated power to the furnace. The crucible temperatures are computer controlled by regulating the current from the aircraft 28 V dc power supply to each of the heaters using voltage-to-current dc amplifiers in the PCU. The thermocouple millivolt output signals are acquired by the data-acquisition unit and stored as temperatures

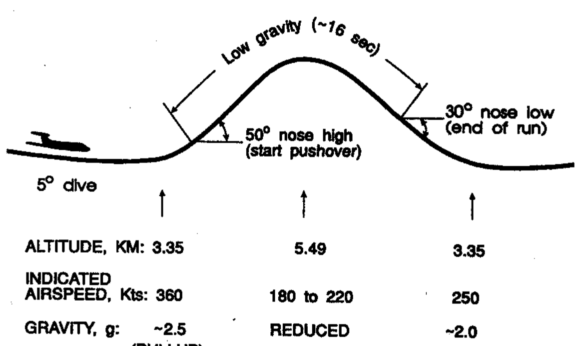

Fig. 1 Typical "low gravity" trajectory ($\pm 0.02g$).

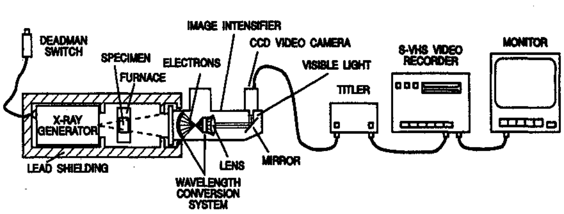

Fig. 2 Real-time radiographic viewing system.

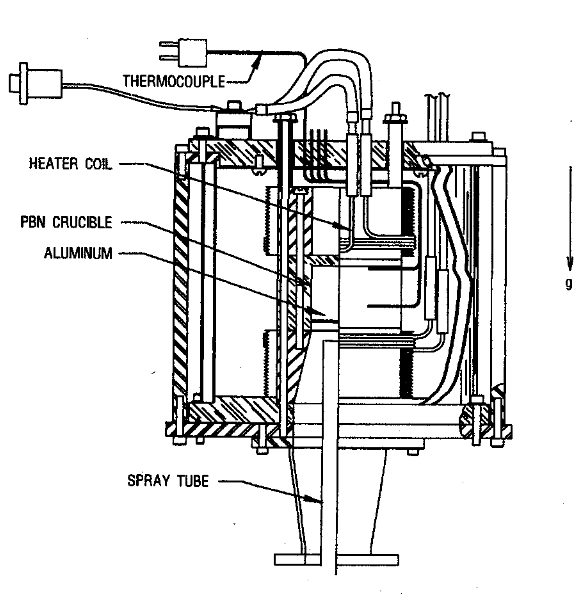

Fig. 3 End-chill directional solidification furnace.

in the computer. The voltage values from the amplifier outputs are determined by the DACS and sent to the computer through the IEEE-488 interface bus. These temperature and voltage values are used to determine the required input power for the furnace and are stored once per minute while heating the furnace and about once per 0.6 s during the quench. The control software calculates and sends an updated voltage signal to the amplifiers in the PCU. The amplifiers, which are directly connected to the appropriate heating coil, adjust their output in proportion to this input signal. The entire system forms a feedback loop allowing control of the furnace temperature. At the appropriate time, the quench is initiated by closing a relay which energizes the pump. This relay is controlled by the computer through an actuator in the data-acquisition unit.

Sample Preparation

Several 99.9999% pure aluminum samples were cleaned in a beaker of acetone solution and placed in an ultrasonic cleaner for 10 min, washed in distilled water, pickled in 10% sodium hydroxide solution for 120 min, and then washed in distilled water and ethanol. Typical sample size was 2.4×2.4× 0.6 cm. The samples were vacuum sealed in 0.063-cm-thick pyrolytic boron nitride (PBN) crucibles using a silver-based vacuum braze material (68.8% Ag, 26.7% Cu, 4.5% Ti).[10] The vacuum chamber pressure was approximately 660 μPa and the braze temperature was 885°C.

Experimental Sequence in Reduced Gravity

Using the computer controlled furnace, the aluminum-PBN crucible system is heated uniformly to 670°C. The typical heat-up rate is 12°C/min. The sample is held at 670°C for several minutes to ensure the aluminum has melted and thermal equilibrium has been reached. During the reduced-gravity portion of the parabolic trajectory, spray quenching is initiated via keyboard input and the real time x-ray system is activated by depressing a deadman switch. This test sequence has been programmed to repeat three times in one flight.

Image Processing

Because the solid/liquid (S/L) interface could only be observed in some of the recorded images during playback of the aluminum solidification, image processing was used to track the S/L interface distance vs time for any desired test run. Figure 4 shows the typical transient movement of the S/L interface. This interfacial movement was essentially independent of the experimental gravity level. Determining the S/L interface required a computer with a frame grabber and an image analysis package. The procedure is described: 1) acquire two live digitized images of a test run, one when the sample is entirely liquid prior to quench, the other when the sample is part liquid and part solid; 2) apply a low-pass filter to each image to remove high-frequency noise; 3) subtract the "all liquid" image from the "part liquid-part solid" image; and 4) stretch the grey scale to magnify the contrast between the light and dark areas. The dark region is solid and the light grey region is liquid. The dark vertical line on the left side of Figs. 4a-4c is assumed to be due to a shift in the two images which are subtracted from each other and is not considered as part of the S/L interface.

Liquid/Vapor Interface Test Conditions

The Bond number was considered in this experiment. The Bond number

$$Bo = \rho_l g r^2/\sigma \quad (1)$$

is a ratio of gravity to surface tension forces. The characteristic length r of the rectangular crucible is one-half of the crucible width, 0.3 cm. The relatively narrow crucible was selected in order to freeze the aluminum in approximately 16 s. This resulted in a Bond number for the experiment of 0.25 in normal gravity.

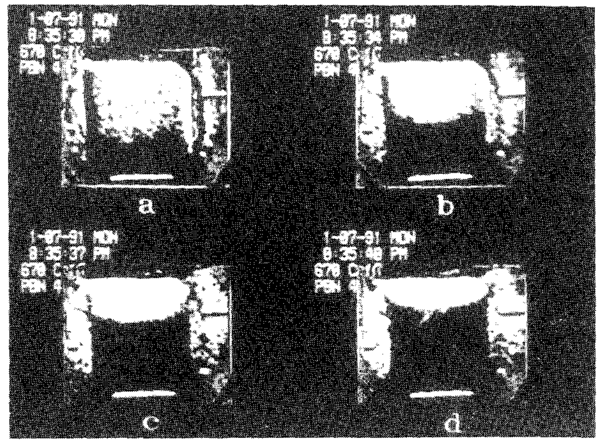

Fig. 4 Tracking the solid-liquid interface of the 2.4×2.4×0.6-cm aluminum sample using image processing. The dark region is solid, and the light grey region is liquid: a) 4 s; b) 8 s; c) 11 s; and d) 14 s after the quench was initiated.

A large change in Bond number about the value $Bo = 1$, i.e., $Bo \gg 1$ to $Bo \ll 1$, can produce significant changes in the free surface curvature. For example, an originally flat meniscus that is gravity dominated ($Bo \gg 1$), can have a radius of constant curvature when surface tension dominated ($Bo \ll 1$) in reduced gravity.[3] Since the Bond number for this experiment is equal to 0.25 in normal gravity, little or no change in the liquid/vapor interface shape is expected in reduced gravity.

Results and Discussion

The curved S/L interface illustrated in Fig. 4 indicates a horizontal temperature gradient was present in the aluminum during the solidification process. This gradient causes natural convective flow in the melt.[11]

The curved S/L interface is a net result of several factors, some of which are cited here. The large amount of latent heat being released (Stefan number, $St = 36.4$) in a rapid manner causes a decrease in the liquid vertical temperature gradient and an increase in the horizontal temperature gradient, thereby increasing the concave curvature of the S/L interface with respect to the melt.[12] The horizontal gradient was also evident based on crucible wall thermocouple data which showed the temperature at 0.23 cm from the boundary adjacent to the advancing S/L interface to be approximately 600°C. Since k_s (217 J/smK) and k_l (94 J/smK) are much greater than k_w (3 J/smK), the interface curvature would be expected to be minimal.[13] However, by k_s being greater than k_l, the S/L interface has a tendency to turn down at the wall, concave with respect to the solid.[14]

The horizontal temperature gradient produces natural convective flow in the melt. The Rayleigh number, based on a characteristic length of 1.27 cm (the horizontal half-length) and a horizontal temperature gradient of 10°C (the bulk temperature of 670°C minus the melting point of 660°C), is $Ra = 1088$ under normal gravity and $Ra = 22$ under 0.02-gravity conditions. A numerical simulation performed by Arnold et al.[13] for GaAs in a gradient freeze furnace showed that the isotherms varied only slightly from those dominated by conduction at $Ra = 164$. This suggests that natural convective flow was occurring in the present experiment. The effect of this convective flow, however, was not evident for the following reasons. First, the rapid average solidification rate of 0.133 cm/s showed no variation with gravitational conditions. Second, there were no dissolved gases to generate bubbles which could become caught in the flow cells; rather the voids originated at the ullage space and did not detach as free floating bubbles.

Figure 5 shows the aluminum at the 70% liquid fill level in the molten state before spray quenching and just after solidification for reduced- and normal-gravity conditions. This Al-PBN system is nonwetting.[15] Since the Bond number is 0.25 at normal gravity, a similar curved liquid/vapor interface occurs for both gravity conditions as shown in Figs. 5a and 5c. In normal and reduced gravity the shrinkage caused void is accommodated by the enlarged vapor volume, as shown in Figs. 5b and 5d, and there is no real difference in the final void shape or location.

Figure 6 shows the aluminum at the 95% liquid fill level. The liquid/vapor interface is curved at both gravity levels, as shown in Figs. 6a and 6c, consistent with the Bond number predictions. Prior to quenching, the molten aluminum is in contact with the crucible lid at both gravity conditions. During solidification in reduced gravity, the aluminum remained attached to the top surface as shown in Fig. 6d. In normal gravity the aluminum remained attached to the top surface until the solid/liquid interface had advanced to about the middle of the crucible. The aluminum then pulled away, resulting in the image shown in Fig. 6b. This particular Al-PBN system was intended to be nonwetting; however, it is assumed that there was some unintentional wetting of the PBN by aluminum on the underside of the lid. This may have occurred during the 885°C vacuum preparation of this Al-PBN crucible system. However, it is not clear why wetting occurred since the Al-PBN system requires about 1000°C in a vacuum for wetting to take place.[15] The results from this test indicate that surface tension forces caused the aluminum to adhere to the crucible lid, creating two voids in reduced gravity, whereas a single large void cavity formed near the lid in normal gravity. In the reduced-gravity test, the ullage volume in the corners enlarged as the sample was solidified. The sample contracted as it solidified, but the aluminum adhered to the lid, resulting in two voids forming instead of one.

At both fill levels, 70 and 95%, no voids were detected at the S/L interface or in the solid aluminum for either gravity level. However, the two prior reduced-gravity experiments that have investigated bubble motion and void formation[5,6] contained dissolved gases which were rejected during solidification and formed bubbles at the growth interface. This produced randomly distributed voids in reduced gravity since buoyancy driven convection was minimized. The degassed aluminum in the present experiment appears to produce fewer voids in reduced gravity, as shown in Fig. 6d, because voids originated only from the initial ullage volume.

The voids were clearly recognizable because the real-time radiographic viewing system provided a clear image of the liquid/vapor interface, and, with the aid of image processing, the solid/liquid interface. Since large voids are of primary interest for space power systems, no post test examinations were made of the samples. The present resolution of 1 mm is considered adequate. Furnace temperatures of 700°C limited the proximity of the heated sample to the image intensifier. The sharpness and resolution of the radiographic images could be improved by cooling the intensifier, thus allowing the furnace and sample to be mounted closer to the intensifier. To the authors' knowledge, this is the first use of a real-time radiographic viewing system to observe solidification in reduced gravity.

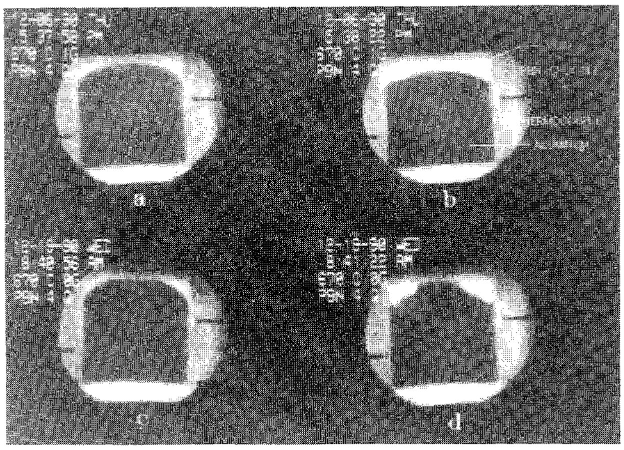

Fig. 6 Void formation during aluminum solidification in normal and reduced gravity for the 95% liquid fill case: a) liquid, normal gravity, prior to quench; b) solid, normal gravity, just after quench; c) liquid, reduced gravity, prior to quench; and d) solid, reduced gravity, just after quench.

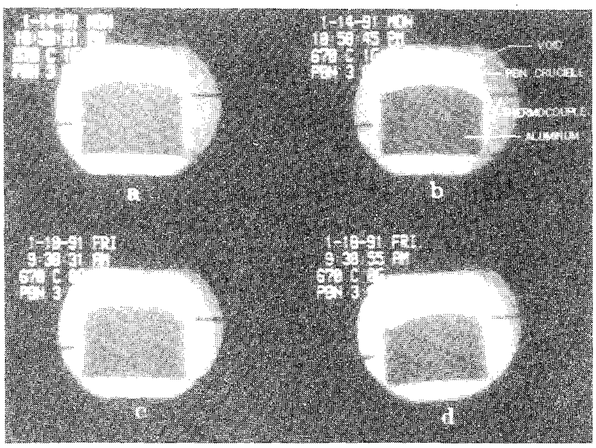

Fig. 5 Void formation during aluminum solidification in normal and reduced gravity for the 70% liquid fill case: a) liquid, normal gravity, prior to quench; b) solid, normal gravity, just after quench; c) liquid, reduced gravity, prior to quench; and d) solid, reduced gravity, just after quench.

Conclusions

Directional solidification of pure molten aluminum in a vacuum sealed PBN container has been observed in both normal and reduced gravity using a real-time radiographic viewing system. Tests in reduced gravity were achieved by flying an aircraft through a parabolic trajectory. The reduced gravity level was $\pm 0.02g$, and lasted lasting approximately 16 s. The typical aluminum solidification rate was 0.133 cm/s in both normal and reduced gravity. Within the crucible a vapor volume was always present. The aluminum-PBN system was surface tension dominated in normal gravity since the Bond number was 0.25. No dissolved gas was intentionally added to the molten aluminum. The conclusions for this system are summarized below:

1) The concave solid/liquid interface indicates a horizontal temperature gradient exists and natural convective flow is present in the melt. However, the rapid freezing rate showed no variation with gravitational conditions, and there were no bubbles in the melt which could become entrained in the flow. Therefore, the effect of convection was not observed to influence the behavior of this aluminum-PBN system.

2) Void formation in a nonwetting crucible is similar in normal and reduced gravity. Because of the material shrinkage (7%), a larger vapor volume at the top of the container resulted. Because the system was dominated by surface tension forces in normal gravity, no liquid/vapor reorientation occurred in reduced gravity.

3) In the case where wetting existed between the molten aluminum and the crucible lid, dominant surface tension forces caused the aluminum to remain attached to the lid in reduced gravity. In normal gravity, the aluminum detached from the lid during solidification. In both cases, volumetric shrinkage was accommodated by the vapor cavity initially present; however, two smaller voids formed in the reduced-

gravity case and only one large void formed during the normal-gravity test.

4) The two previous reduced-gravity experiments that have investigated bubble motion and void formation[5,6] contained dissolved gases, resulting in a random void distribution in reduced gravity. The aluminum used in the present experiment did not contain dissolved gases and yielded fewer voids, which originated only at the initial ullage volume.

5) To the authors' knowledge, this is the first use of a real-time radiographic viewing system to observe solidification in reduced gravity. This system produced images with 1-mm resolution, a clear liquid/vapor interface, and with the assistance of image processing, the solid/liquid interface.

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