mocouples were located on the disks.

The dimensions of the liquid bridge are essentially governed by the Bond characteristic length $L_b = \sqrt{\sigma/\rho g}$, where ρ is the density of the liquid and g is the intensity of the (residual) gravity vector. For most liquids of interest L_b, on the earth, is of the order of 10^{-1} cm ($L_b = 0.149$ cm for silicone oil) (*5*). If R denotes the smallest radius of curvature of the interface, hydrostatic equilibrium (*1*) and stability (*6*) analyses lead to the conclusion that both L (for g directed along the axis of the liquid bridge) and R can be at most of the order of L_b. Since L_b is scaled as the inverse square root of the gravity level, it follows that in the nominal Spacelab environment ($g/g_0 = 10^{-4}$) one can obtain liquid bridges two orders of magnitude higher than on the earth.

This has been confirmed by the Spacelab experiment. With a disk diameter of 6 cm, flow patterns of the type shown in Fig. 1 were observed in columns as high as 6.8 cm. Columns were formed by injecting liquid from a reservoir through a hole in the feeding disk and concomitantly increasing the distance between the two disks. After a column had been kept in isothermal conditions for a time, it was observed that it reached a truly quiescent state: all tracers were at rest. Power was then switched on and tracer movement was immediately triggered. Thus the flow was proved to be a Marangoni flow. This also shows that, at least during this experiment, the gravity level was less than 10^{-3}, otherwise the liquid column would have collapsed. By comparison, simulations on the ground could be performed only with liquid bridges having a radius of 3 mm and heights of the same order.

The momentum produced at the interface by the Marangoni force diffuses into the liquid. Let l be the nondimensional penetration depth for momentum diffusion, measured normal to the interface and referred to the column height, L. Flow regimes depend on the order of magnitude of l, which in turn depends on the order of magnitude of the conditional Reynolds number $R_m = V_m L/\nu$ (*4*, pp. 349–358), where ν is the kinematic viscosity of the liquid, $V_m = [\sigma(T_c) - \sigma(T_H)]/\mu$, the Marangoni speed, and μ is the dynamic viscosity.

For R_m less than or equal to one, the penetration depth is on the order of one and the flow regime is of the Stokes type (inertia forces negligible, $R_m \ll 1$) or the Navier-Stokes type [inertia and viscous forces of the same order of magnitude, $R_m = O(1)$]. For $R_m \gg 1$, l is much less than one and momentum diffusion from the interface into the liquid is confined to a thin layer, which we call the Marangoni boundary layer (*7*), whose nondimensional thickness δ (referred to L) is on the order of $R_m^{-1/3}$. Everything else being the same (fluids, geometry, and imposed temperature difference) in microgravity environments, the conditional Reynolds number R_m may be considerably larger than on the earth because of the much larger extension of the liquid bridge. Hence, Marangoni boundary layers are very likely to be established and features of the flow will be substantially different from those in ground simulations of Marangoni convection. This scale effect provides additional evidence of the need for experimentation in space.

Attainment of a Marangoni boundary-layer regime in the Spacelab experiment has been assessed by estimating the order of magnitude of the maximum flow velocity, V_r, from the available data. The order of magnitude of V_r in Marangoni convection depends on the flow regimes (*1, 3*). In the absence of Marangoni boundary layers, V_r is of the same order as the Marangoni characteristic speed, V_m. When boundary layers are present V_r is of the order of $V_m\delta$ and is thus substantially smaller. Estimates of V_r indicated that Marangoni boundary-layer regimes were attained in most of the runs.

L. G. Napolitano
Institute Umberto Nobile, University of Naples, I-80125 Naples, Italy

References and Notes

1. L. G. Napolitano, paper presented at the 2nd Levitch Conference, Washington, D.C., November 1978.
2. ——, *Acta Astronaut.* **6**, 1093 (1979).
3. ——, *ibid.* **9**, 199 (1982).
4. ——, in *Material Sciences in Space* (ESA-SP-142, European Space Agency, Paris, 1979), pp. 289–296.
5. Techno System, "Study on fluid phenomena influencing the design of zero-*g* experiments—final report," (Techno System Rep. TS-1-80, Naples, 1980).
6. W. Heywang, *Z. Naturforsch.* **32**, 238 (1956).
7. L. G. Napolitano, in *Material Sciences in Space* (ESA-SP-142, European Space Agency, Paris, 1979), pp. 349–358.
8. I acknowledge the financial support of the Italian Space Plan (PSN-CNR) and the contributions of those who collaborated in the different phases of the investigation: R. Monti, C. Golia, and G. Russo. The entire 1ES328 team expresses deep appreciation for the valuable and creative work of the two payload specialists: B. Lichtenberg and U. Merbold.

27 March 1984; accepted 23 May 1984

Solidification and Ostwald Ripening of Near-Monotectic Zinc-Lead Alloys

Abstract. *Studies of melting and resolidification were carried out on Spacelab 1 on the zinc-lead binary, an alloy which exhibits a miscibility gap in the liquid state. The possibility of maintaining the state of homogeneous dispersion of lead in the zinc matrix in a microgravity environment was verified. The second objective of the experiment was to study Ostwald ripening of the lead droplets (manifested as slow coarsening of the droplets) within the region of immiscibility. An increase in droplet size was observed and may have been due to Ostwald ripening, although the effect on droplet size of precipitation during cooling must be analyzed before this can be determined conclusively.*

The Zn-Pb binary is representative of the large group of alloy systems that exhibit a miscibility gap in the liquid state. At high temperatures molten zinc dissolves lead completely, but below a critical temperature the liquid separates into two phases, forming a suspension of lead-rich droplets in a zinc-rich liquid matrix. Under terrestrial gravity, the lead droplets sink to the bottom, causing complete demixing of the system. Under zero gravity and in the absence of convection, such suspensions should be stable for extended times. This was demonstrated for the model system water-oil in a Skylab experiment (*1*). For immiscible metallic systems, however, coarse demixing has occurred in nearly all microgravity experiments so far, including one Apollo-Soyuz experiment on Zn-Pb (*2*). This was attributed to Marangoni convection, residual acceleration, or unspecified "special effects."

One objective of experiment 1ES313 on Spacelab 1 was to verify the possibility of maintaining the state of homogeneous dispersion in Zn-Pb alloys and of bringing the dispersion back to the solid state without disturbance by particles being pushed ahead of the solidification front. Terrestrial experimentation (*3*) had shown that, at the solidification rate envisaged for the Spacelab experiment, the lead particles would not be pushed ahead.

The second and main objective of our Spacelab 1 experiment was to study Ostwald ripening of the lead droplets within the region of immiscibility. This would manifest itself as a slow coarsening of the droplets, driven by the tendency toward minimum total interfacial energy in the system. On the earth this process is disturbed by gravity separation; additional disturbance could result from density-driven or surface energy–driven convection causing collisions and coalescence of droplets. (To minimize such disturbances, we chose a system with a low volume fraction of small droplets.) The sustained microgravity of Spacelab 1 would offer a unique chance to study Ostwald ripening in liquid-liquid systems.

To test the existing theory of Ostwald ripening (*4, 5*) from first principles, one would need a series of isothermal hold times, all long and preferably at two different temperatures. The limitations of the first Spacelab mission allowed only two short runs, and the second run had to be canceled because of an earlier furnace malfunction. It was clear that, within the available time, measurable ripening effects could be expected only for extremely small droplets. Therefore, instead of varying the hold time, a series of different initial states would be used by incorporating specimens with different amounts of lead in each furnace run. The lead particle size in these specimens would be measured before the flight and compared to measurements after the flight; corrections would be made for shrinkage of the particles by dissolution in the liquid matrix during heating up to the isothermal hold, and for their growth by reprecipitation of the dissolved material during subsequent cooling.

Experimental procedure. Zinc-lead melts were homogenized in an induction furnace, degassed, and quench-cast into a cooled cylindrical copper mold 6 mm in diameter. Quenching at about 200 K per second produces a homogeneous distribution of lead particles in the zinc matrix. The particles are so fine that they can be studied only in a scanning electron microscope. To contain the vapor pressure of the zinc even in case of uncontrolled overheating, the Zn-Pb specimens were enclosed in thick-walled, electron beam–welded tantalum tubes. Six such tubes, each containing three slugs of alloy, were placed in a standard cartridge of the isothermal heating facility of Spacelab. The alloy compositions ranged from 2 to 6 percent lead by weight.

Figure 1 shows the relevant part of the miscibility gap. A hold temperature of 450°C was planned, to preserve undissolved lead droplets for ripening in alloys containing ≥2 percent zinc by weight. Despite careful preflight testing in a duplicate of the isothermal heating facility at Deutsche Forschungs- und Versuchsanstalt für Luft- und Raumfahrt

Table 1. Mean diameters of lead particles in solidified specimens.

Time	Particle size (μm) in specimen with Pb content of			
	2 percent	2.5 percent	3.5 percent	4 percent
Before flight	1.1	1.4	1.2	1.2
After flight	13.5	12	11	12

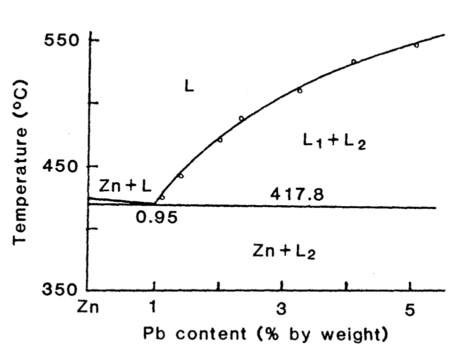
(DFVLR) in Cologne, the temperature plateau in flight was about 25 K higher (Fig. 2). In addition, there was some overshooting of the cartridge temperature before it settled on the plateau. Unfortunately, the amount is not known because of a gap in data transfer during the critical period. We have to assume that the specimens containing 2 and 2.5 percent lead were almost or fully homogenized.

Preliminary results. Up to now we have investigated samples with 2 to 4 percent lead by weight. The overheated samples with 2 and 2.5 percent lead, being the least valuable, were evaluated first.

No macroscopic segregation or phase separation was observed in any of the samples; all specimens showed a homogeneous dispersion structure. The lead particles were much larger than before the flight (Table 1). For the samples with 2 and 2.5 percent lead, complete particle size distributions have already been measured (Fig. 3); for the others, this remains to be done.

Discussion. The large particle size in the overheated samples is probably due to renucleation of droplets on entering the region of immiscibility at a very slow rate, or to precipitation on a few droplets that may have survived the temperature overshoot.

In the specimens with 3.5 and 4 percent lead, enough lead droplets should have remained undissolved to exclude independent renucleation. The coarsening of the droplets could have been caused by several processes: Ostwald ripening; dissolution of all particles below a critical size during heat-up and overshoot, which would concentrate all



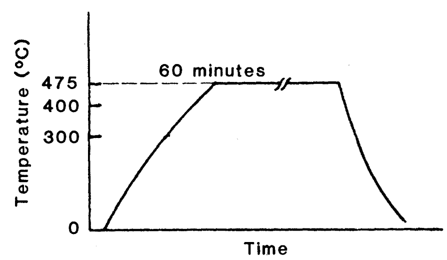


Fig. 1 (left). Zinc-lead binary system near the monotectic region. Fig. 2 (right). Temperature-time profile during the Spacelab experiment.

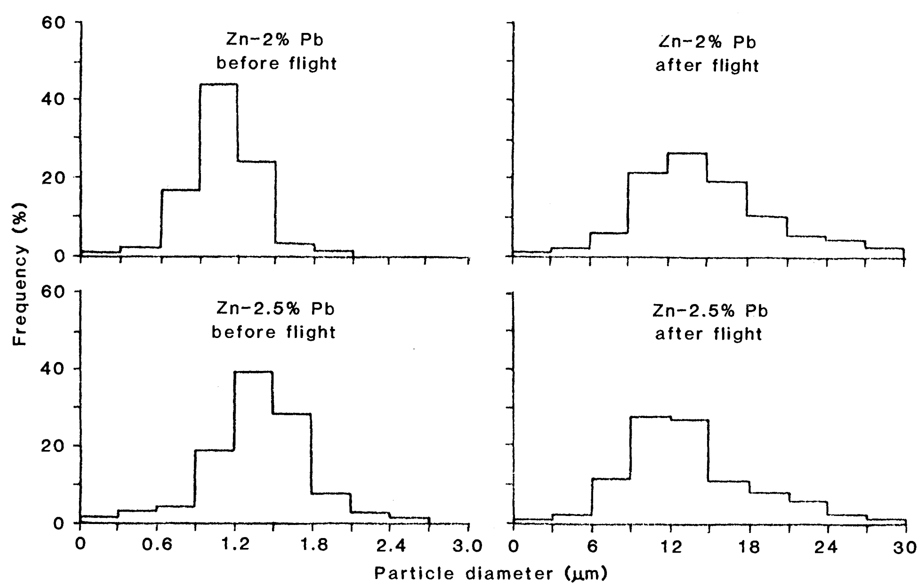


Fig. 3. Frequency distribution of particle diameters for several samples.

reprecipitation to the remaining droplets during subsequent cooling; or coagulation due to droplet collisions induced by Marangoni convection.

We will first test the hypothesis of convection-induced coagulation. The velocity, v, of a particle of radius r is given by

$$v = -\frac{2}{3}\frac{r}{2\eta_1 + 3\eta_2}\frac{d\sigma}{dT}\frac{dT}{dy} \qquad (1)$$

where η_1 is the viscosity of the zinc matrix and η_2 is the viscosity of the lead droplets. The temperature coefficient of surface tension, $d\sigma/dT$, is of the order of 0.1 mJ/m^2-K between L_1 and L_2 in a miscibility gap system (*6*). Assuming a temperature gradient, dT/dy, of the order of magnitude of 1 K per centimeter, we obtain velocities between 0.1 and 2 μm/sec for particle sizes from 1 to 20 μm. These particle velocities are about two orders of magnitude smaller than those tested in earlier terrestrial experiments, which could be shown to make a negligible contribution to particle coarsening (*3*).

For Ostwald ripening, the LSW theory (*4, 5*) predicts the following relation between mean particle radius and time:

$$\bar{r}(t)^3 - \bar{r}(0)^3 = kt \qquad (2)$$

$$k = \frac{8\sigma C_0 D \Omega^2}{9RT} \qquad (3)$$

The boundary energy, σ, between the liquid lead-rich particles and the zinc-rich melt is about 100 mJ/m^2 (*7*); the solubility, C_0, of lead in molten zinc is 2 percent by weight at 480°C (see Fig. 1); the molar volume, Ω, of lead is 18 cm^3/mole; and the gas constant, R, is 8.3 J/mole-K. The rate constant, k, of Ostwald ripening calculated from the mean particle radius before and after flight is 0.035 μm^3/sec, which would yield a diffusion coefficient for lead in liquid zinc at 480°C of 10^{-4} cm^2/sec. This result is in agreement with a value for the diffusion of lead in liquid zinc in a 1-*g* experiment by Pelzel [$D_{500°C} = 3 \times 10^{-4}$ cm^2/sec (*8*)], but is about 30 times larger than values that were obtained in our terrestrial Ostwald ripening experiments (*9*). While Pelzel's data may have been high because of convection, ours may have been low because of loss of large particles by sedimentation. In view of the lack of diffusivity measurements unaffected by gravity, we can only note that the diffusivity calculated from our Spacelab experiment is within the range spanned by available data.

The discussion above shows that Ostwald ripening would be a plausible mechanism for the observed increase in droplet size. However, the effect of precipitation on the droplets during cooling from the hold temperature has to be analyzed more accurately before final conclusions can be drawn. For this, the number of droplets per unit volume must be determined from an accurate size distribution analysis, and this will be among the next steps in the evaluation of our Spacelab results.

ALBERT KNEISSL
Institut für Metallkunde und Werkstoffprüfung, Montanuniversität A-8700 Leoben, Austria
HELLMUT FISCHMEISTER
Max-Planck-Institut für Metallforschung, D-7000 Stuttgart 80, Federal Republic of Germany

References and Notes

1. L. L. Lacy and G. H. Otto, AIAA paper No. 74-1242 (New York, 1975).
2. C. Y. Ang and L. L. Lacy, *ASTP Experiment MA-044, NASA TMX-64956* (Marshall Space Flight Center, Huntsville, Ala., 1975).
3. A. Kneissl, R. Pfefferkorn, H. Fischmeister, in *Proceedings of the 4th European Symposium on Materials Sciences under Microgravity, Madrid* (ESA SP-191, European Space Agency, Paris, 1983), p. 55.
4. I. M. Lifshitz and V. V. Slyozov, *J. Phys. Chem. Solids* **19**, 35 (1961).
5. C. Wagner, *Z. Elektrochem.* **65**, 581 (1961).
6. H. Fredriksson, in *Proceedings of a Workshop on the Effect of Gravity on Solidification of Immiscible Alloys, Stockholm* (ESA SP-219, European Space Agency, Paris, 1984).
7. R. Pfefferkorn, thesis, Montanuniversität Leoben, Leoben, Austria (1981).
8. E. Pelzel, *Z. Metallkd.* **61**, 289 (1970).
9. A. Kneissl and H. Fischmeister, *Metall (Berlin)* **37**, 131 (1983).
10. Financial support by the Austrian Ministry of Science and Research is gratefully acknowledged.

27 March 1984; accepted 30 May 1984

Unidirectional Solidification of Cast Iron

Abstract. *The segregation of sulfur in liquid cast iron was studied under conditions of microgravity on Spacelab 1. A rod of cast iron containing carbon, silicon, sulfur, and phosphorus was unidirectionally solidified at four different rates. The influence of sulfur on the graphite structure and the stability of an aluminum oxide skin deposited on the surface of the specimen were investigated.*

On day 4 of the Spacelab 1 mission experiment 1ES325 was conducted as planned. Liquid cast iron containing 3.9 percent carbon, 1.5 percent silicon, 0.025 percent sulfur, and 0.01 percent phosphorus was unidirectionally solidified at four different rates varying between 0.1 and 1 mm/min (*1*). Before the successful run of the experiment a problem occurred with the temperature control of the isothermal heating furnace (IHF) with gradient device after it had been preheated. After the furnace had been cooled down, the experiment was restarted by the payload specialists.

Analysis. The specimen as received from Spacelab after the mission was examined by different techniques. It consisted of a cast iron rod 139 mm long contained in a crucible. An Al_2O_3 skin was deposited on the surface of the specimen (*2*). The examination proceeded from the outside to the inside. First, x-rays were used to determine the location and position of the cast iron within the crucible. In addition, the condition of Pt-PtRh10 thermocouples mounted at the surface of the rod within the crucible was established.

The crucible was then removed with a rapidly rotating diamond saw, which made two cuts along the length of the crucible with a depth equal to the thickness of the crucible. After removal of the crucible, the Al_2O_3 skin was examined by light optical microscopy and scanning electron microscopy combined with x-ray energy-dispersive analysis. From this, information was obtained on the morphology and chemical composition of the skin.

Then the rotating diamond saw was used to cut the sample over its whole length into two parts. For this purpose the sample was moved along the cutting wheel with a speed of about 0.5 mm/min.

After grinding on silicon carbide abrasive paper and polishing on diamond with particle sizes of 3, 1, and 0.25 μm, together with intermittent weak nital etching, the graphite structure was studied with a light optical microscope.

Results and discussion. The x-ray photography showed that the liquid column of the specimen had been separated into two parts, which were 85 mm and 26 mm in length. Two effects may have played a role in this separation. The highest temperature in the IHF was in a confined region near the end of the rod. This caused the material to start melting in a radial direction at some distance from the outer end of the rod. Unlike most metals, gray cast iron shrinks during melting. Because of this shrinkage and the surface tension, the first melted zone had a tendency to break into two parts under microgravity conditions. Furthermore, the unexpected development of some gases may have caused the smaller part to be pushed toward the end of the crucible as some pressure built up. This effect could also explain why some of the molten iron may have been pushed