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MEMORANDUM

NASA TM 78144

MIXING OF TWO LIQUID METALS ON SPAR PAYLOAD DUE TO SPIN-UP AND SPIN-DOWN

By Charles F. Schafer
Space Sciences Laboratory
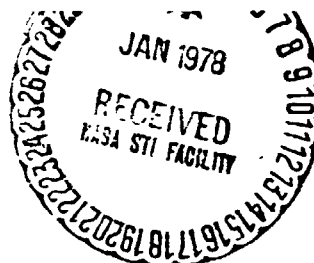
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16. ABSTRACT

The work described was motivated by unexpected results from SPAR experiments directed at obtaining fine dispersions of metal systems that in a 1-g environment tend to segregate very rapidly. The SPAR samples exhibited segregation also. The physical processes occurring in fluid samples, such as the SPAR samples before solidification, are shown to be insufficient to produce a well-mixed liquid by the time solidification was initiated. This would result in solidified samples with the type of segregation noted in the SPAR samples. Experimental evidence and analytical arguments are presented.

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INTRODUCTION

The Space Processing Applications Rocket (SPAR) Project provides experimenters with the opportunity to perform materials science and fluid dynamics experiments in approximately 5 min of low-g. This low-g environment has been demonstrated to be typically in the 10^{-5} g or lower range for SPAR I[1] [1]. A SPAR flight consists of a spin-stabilization phase beginning with launch, a rapid spin-down, a low-g coast phase, and a reentry phase. The rotation rate is shown as a function of time for the SPAR II flight in Figure 1. The $\leq 10^{-5}$ g accelerations occur during the coast phase, and these low levels have been attributed to residual spin and precession of the payload after despin occurs.[2] Most experiments performed onboard SPAR payloads depend on quiescent fluid conditions during this low-g period, and, therefore, it is critical that any motions induced by the launch procedure be damped to near zero before the experiments are begun. It was recognized early in the SPAR program that this was the case, and a study was conducted to determine the necessity of a nonspin platform to preclude these spin effects. Results from studies conducted at the Marshall Space Flight Center[3] showed that fluid motions resulting from the spin-up/spin-down processes would damp out in a few seconds for fluid systems considered typical of those to be flown onboard SPAR. These studies demonstrated that the nonspin platform was not necessary for the SPAR flights. The payloads themselves then experience the spin-up and spin-down profile shown in Figure 1. Experiments must be designed so that this spin profile does not deleteriously affect results. For example, some experiments

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1. Holland, R. L.: Acceleration Levels for Space Processing Application Rockets (SPAR). NASA Technical Memorandum to be published.

2. Ibid.

3. Personal communication with L. L. Lacy, Space Sciences Laboratory, Marshall Space Flight Center.
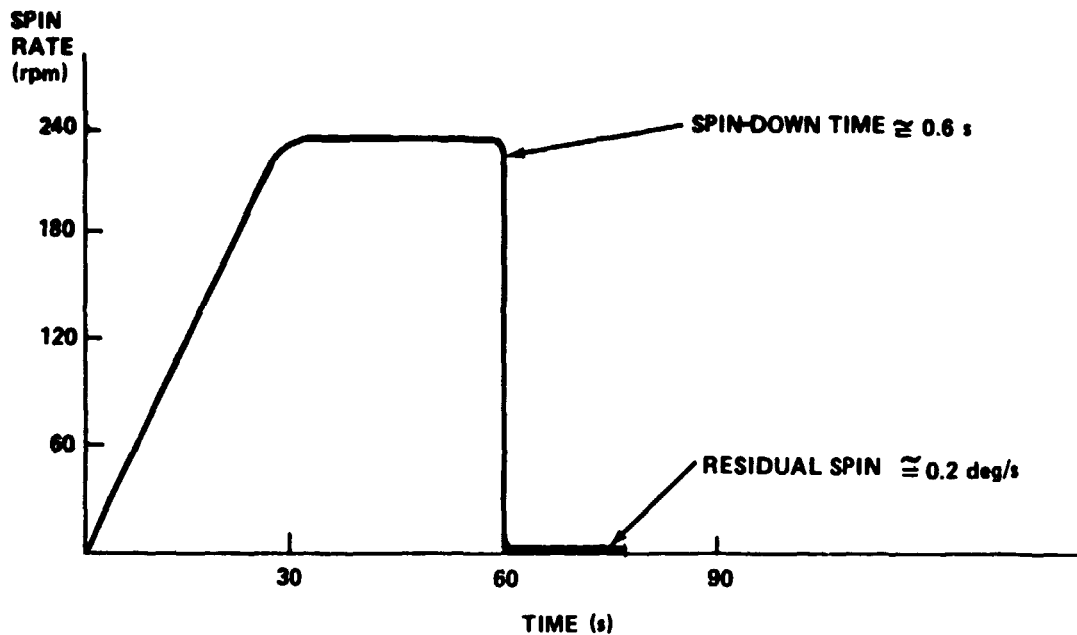
Figure 1. SPAR II spin profile.

require that the experimental liquid be in a solid phase until after despin, while other experiments depend on viscous damping to still fluid motions [2]. Still other experiments attempt to use the spin profile to effect homogenization in liquid multicomponent systems [3]. It is the unexpected results of these experiments [3] which motivated the work described in this report.

SPAR II IMMISCIBLE SYSTEMS EXPERIMENTS

Two experiments flown on SPAR II were aimed at producing fine scale dispersions of the two components of a metal-metal system for which these dispersions are achievable 'n bulk form in a 1-g environment. These materials exhibit behavior characterized by a liquid phase "miscibility gap" (Fig. 2). That is, the two metals are completely soluble above the liquidus line, but a gap exists between the liquidus and solidus lines in which the two metals are in equilibrium with each other as separate immiscible liquids. Slow cooling (so that near equilibrium conditions are maintained) from above the miscibility gap then should proceed from a homogeneous liquid phase alloy to a state in which
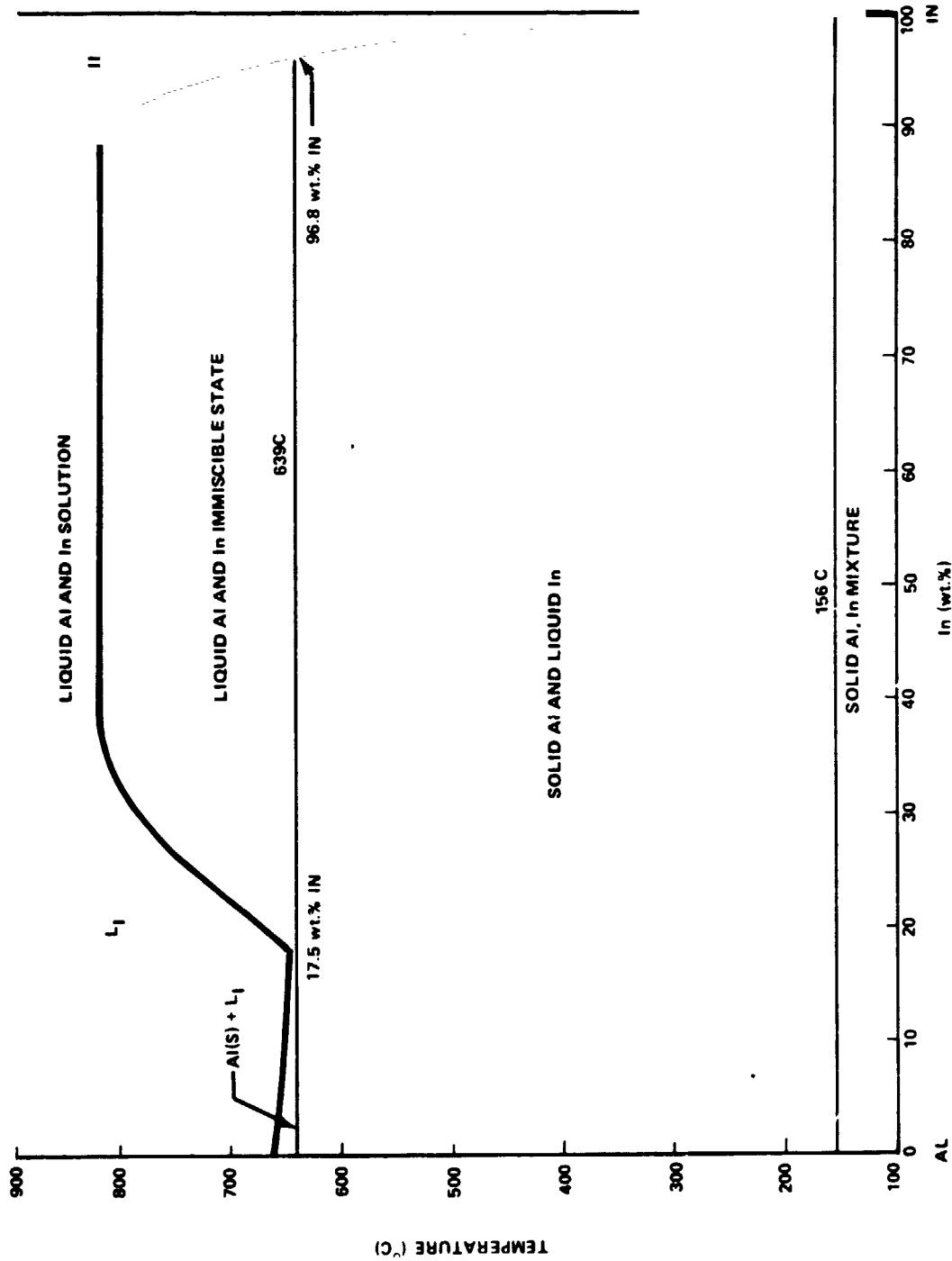
2

Figure 2. Al–In equilibrium diagram (Metals Handbook [4] modified by Gelles [3]).

the preferred (from energy considerations) configuration is a fine dispersion of
one liquid in the other effected by molecular diffusion. This dispersion will
become coarser due to various mechanisms, including Ostwald ripening, fluid
convection effects yielding collisions among the droplets, and sedimentation when
the system is in a gravitational field. The final configuration will be dictated
by minimization of free energy (surface energy) and will be characterized by
complete segregation of the two liquid metals in a manner governed by the state
of containment of the metals and the presence or absence of a gravitational field.
Figure 3 shows final configurations expected for a variety of conditions [3].
When these procedures are followed in a 1-g environment, sedimentation proc-
esses very rapidly segregate the two liquid metals as they become immiscible
so that no finely dispersed solid phase can be achieved in bulk form. Very high
quench rates are accessible using splat cooling techniques, but only fine wires
or ribbons of solid material can be obtained. To obtain these fine dispersions
in bulk form, the melt must be solidified in the absence of sedimenting forces
and rapidly enough that Ostwald ripening and other processes do not coarsen
the dispersion too much. That is, a low-g experiment must be used with rapid
quench rates. With this understanding of the problems associated with preparing
fine dispersions from metallic systems characterized by a liquid state miscibility
gap, the experiments mentioned previously were designed for performance in
the SPAR flight series. The experiments used approximately 1 cm^3 samples
composed of various ratios of indium (In) and aluminum (Al). (The phase diagram
is shown in Fig. 2.) The operational procedure [3] was designed under limita-
tions of total furnace on-time (due to waste heat extraction considerations). It
consisted of a 15-min presoak period at 950°C, which is above the In-Al misci-
bility gap, holding that temperature until approximately 154 s after launch
(through spin-up and spin-down), and then initiating a rapid quench which
carried the system through the miscibility gap to complete solidification in less
than 15 s.

The processed samples were returned to the principal investigators for
metallurgical analysis. The results of the analyses by Gelles are reported in
Reference 3. The flight sample yielded results different from ground-processed
samples, but did not yield the expected results. Ground-based samples carried
through the same procedure evidenced nearly complete segregation of the In-rich
and Al-rich regions. There was no evidence of any gravitational-type segrega-
tion, but the Al-rich phase was surrounded by the In-rich phase (Fig. 4).
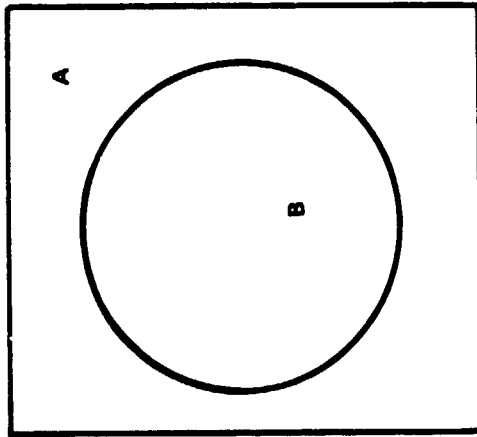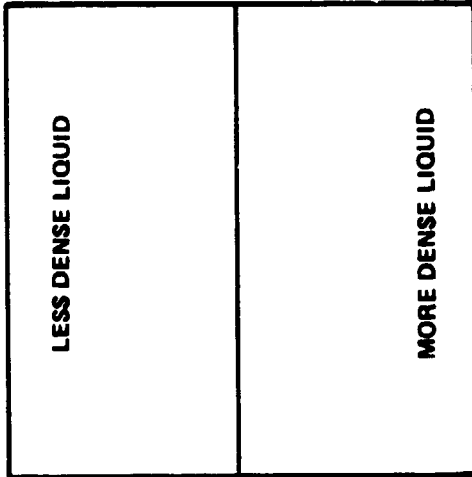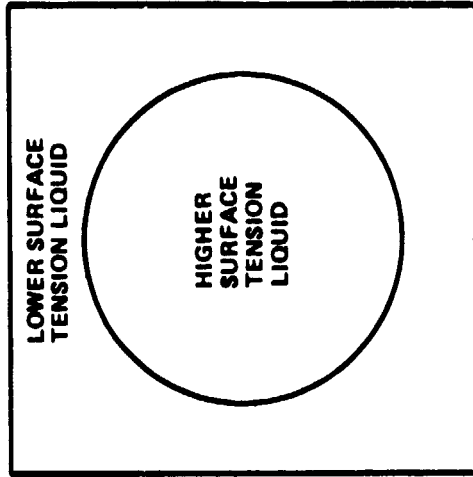
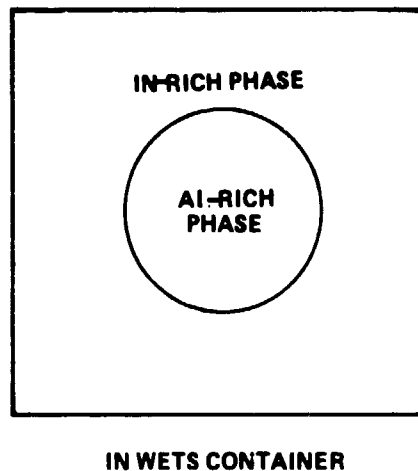Figure 3. Equilibrium configurations of immiscible liquids.

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Figure 4. Post-flight sample configuration,
Experiment 74-30.

HYPOTHESES TO EXPLAIN SPAR II IMMISCIBLE
SYSTEMS EXPERIMENT RESULTS

Two possible explanations have been proposed to account for the afore-
mentioned observations. First, kinetic effects which occur after the rapid
cooling of the sample is initiated have been proposed. These effects must take
the system from a completely interdiffused alloy (at a temperature above the
miscibility gap) to a nearly completely segregated system in a time of less
than 15 s (the time required to solidify the sample from a temperature above
the miscibility gap). The second possible explanation for these results is that
the two materials were not completely mixed at the time cooling was initiated.
This could have resulted from either failure to heat the materials above the
miscibility gap or from the inadequacy of the operational procedure to produce
a homogeneous system. The work described in this report attempts to deter-
mine the most probable of these possible causes of the observed results.

The separation of materials from a homogeneous state as discussed
here requires, first, that diffusion act, producing a dispersion of the separate
materials. Since the experiment time is on the order of 10 s (for cooling to
solidification from above the miscibility gap), one needs to find a process or
processes which can almost completely separate the materials in that amount
of time. Considering diffusion first, it is found that for a diffusion coefficient

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of 10^{-5} (cgs units) the following size droplets can be formed in about the times shown:

$$10 \, \mu m \rightarrow 10^{-1} \, s$$

$$30 \, \mu m \rightarrow 1 \, s$$

$$100 \, \mu m \rightarrow 10 \, s \ .$$

It can be seen that droplets on the order of 10's of micrometers in diameter can be formed by diffusion in times less than 1 s and that considering diffusion to act over the entire experimental period does little to affect macroscopic segregation. It may be assumed that any segregation on the scale of the sample size must begin with a diffusional process which acts nearly instantaneously compared to total experiment time to produce a fine dispersion of droplets ($\sim 10 \, \mu m$ or less) and then culminate in some macroscopic kinetic process. In a 1-g field, the dominant kinetic effect would be gravitational settling. If one assumes a dispersion of droplets on the order of 30 μm in diameter and applies Stokes' law to determine fall velocities,

$$V_f = \frac{1}{18} \frac{g d_s^2}{V} \left(\frac{\rho s - \rho}{\rho} \right) \approx 0.2 \, cm/s \quad ,$$

it can be seen that a sample on the order of 1 cm in size would undergo complete segregation in an experiment lasting as long as 10 s. However, when "g" is reduced by 4 to 6 orders of magnitude, as in a typical SPAR expe: ment, Stokes settling becomes insignificant. Further, any kinetic mechanism that can be found which might affect the observed segregation must produce velocities in the experimental fluid on the order of 0.1 cm/s (actually, a velocity differential between the droplets of one material and its host medium must exist).

The simplest explanation of the results obtained in the SPAR immiscible material experiments is that a homogeneous solution of In and Al did not exist at any time. It has been demonstrated that the temperatures obtained during the experiments were sufficiently high (Fig. 5) to be above the miscibility gap shown in the In-Al phase diagram. This suggests then that the procedure for obtaining a homogeneous solution was inadequate. This procedure can be analyzed step by step to determine the most probable configuration of the In-Al samples before
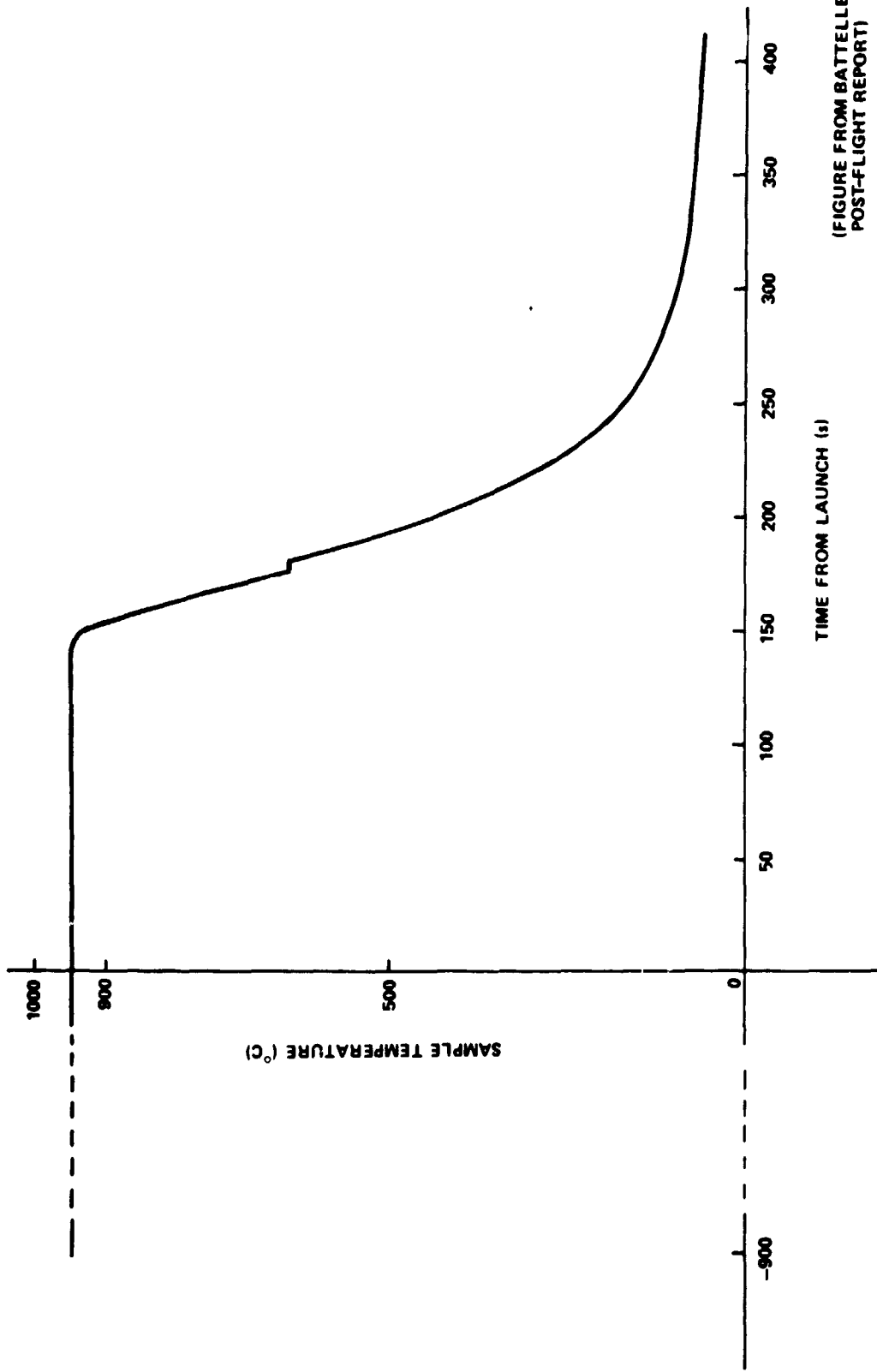
Figure 5. Thermal profile, Experiment 74-30.

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quench was initiated. First, the samples are positioned in the SPAR furnace with the more dense $(d \approx 7.0 \text{ gm/cm}^3$ in liquid form) In above the Al $(d \approx 2.4 \text{ gm/cm}^3$ as a liquid). When melted, the In will flow to the bottom of the container, with the lighter Al floating on it. Very little mixing should occur during this process. The system is then heated above the miscibility point and held for 15 min before launch. Because of the large density difference between In and Al, this configuration will be highly stable with respect to convection; that is, mixing will be limited to diffusion processes. Progress of a diffusion front during this time will be only approx ately 1 mm (approximately 20 percent of sample length) assuming a diffusion coefficient of 10^{-5} (cgs units). The configuration of the samples immediately before launch should be approximated by the sketch shown in Figure 6. The launch begins a spin-up phase from 0 to 240 rpm in approximately 30 s. This 240 rpm is maintained for another 30 s. This spin-up is not expected to significantly affect the mixing process. At the lower spin rates, the density stratification will tend to reduce the effects of the Ekman boundary layers in promoting mixing [5]. At the higher spin rates, the sample can be quickly dominated by centrifugal effects which can even more strongly stratify the fluid system (Fig. 7). If the sample is located 2 cm from the spin axis, the centrifugal acceleration will be approximately 1.5 g. After thruster burnout, the fluid configuration should be approximated by Figure 8. Diffusion between the In and Al (predominately) layers should be
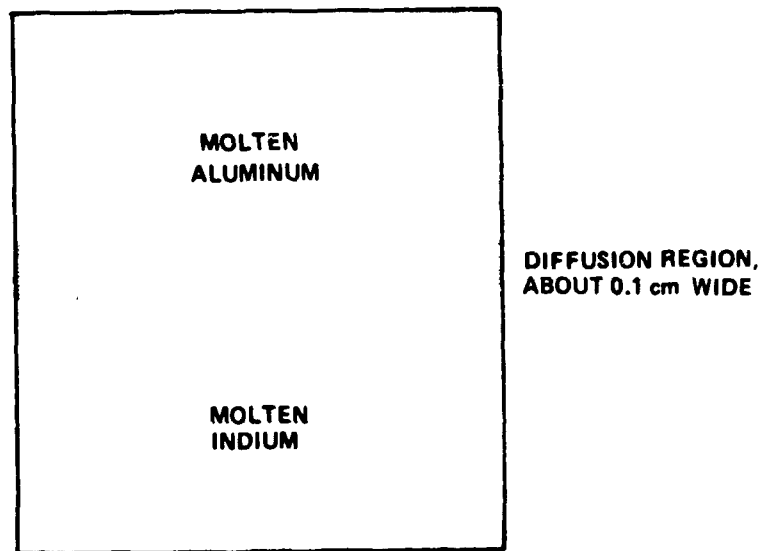
MOLTEN
ALUMINUM

DIFFUSION REGION,
ABOUT 0.1 cm WIDE

MOLTEN
INDIUM

Figure 6. Sample configuration at time of launch,
Experiment 74-30.
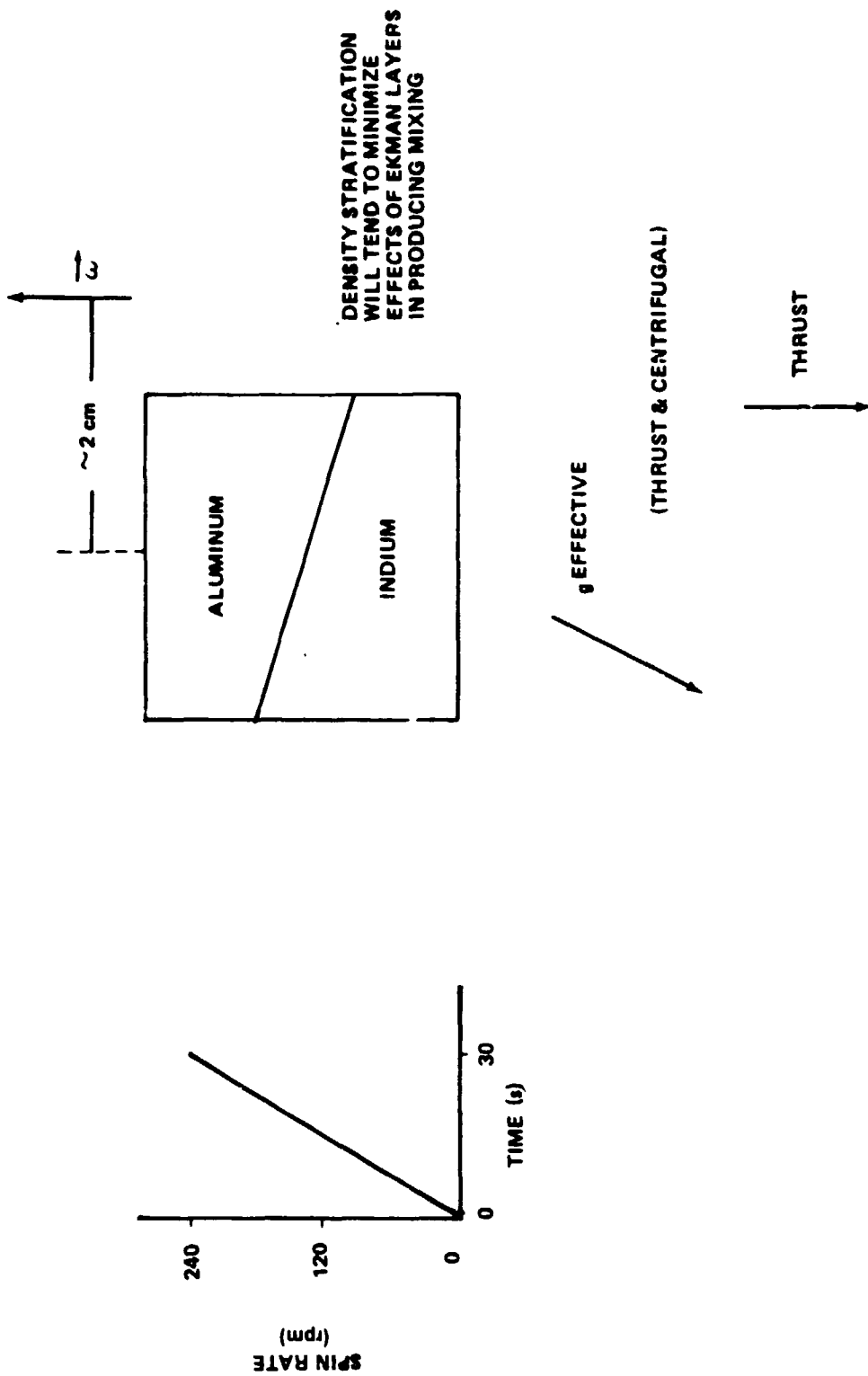
Figure 7. Sample configuration during spin-up (rocket liftoff to burnout).

10

g EFFECTIVE →

$\uparrow\vec{\omega}$

INDIUM   ALUMINUM
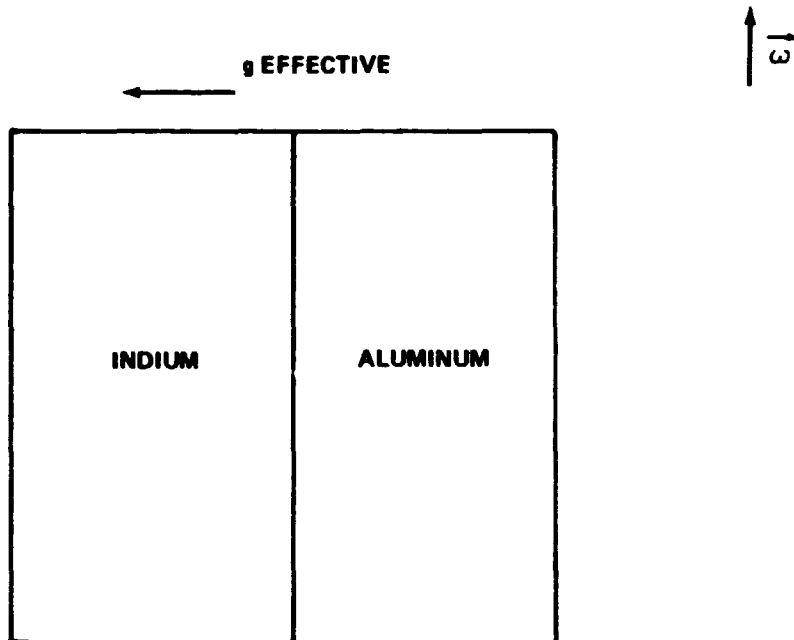
SAMPLE OFFSET ABOUT 2 cm FROM AXIS OF ROTATION

Figure 8. Sample after spin-up (no rocket thrust).

the dominant mechanism for mixing, and this cannot produce a significant amount in the 1 min of spin-up time. At the end of the 60 s spin-up, there is a deployment of "yo-yo" mechanisms by the rocket payload which reduces the spin rate to very low levels (~ 0.1 deg/s) by changing the moment of inertia about the longitudinal axis. This despin is accomplished in approximately 1 s. It is expected that the liquid metal samples will have been in solid body rotation before despin and that despin will result in turbulent motion which should persist for only a few seconds. The time scale for spin-down (linear effects) is given by Greenspan [5] as $t = (L^2/\Omega \nu)^{1/2}$, where L is a characteristic dimension, Ω is the rotation rate, and ν is the kinematic viscosity. For the case discussed in this report, $t \approx 6$ s. Since this time scale is for linear effects, the turbulent case that can be expected here should have an even shorter time scale. (This spin-down time scale is confirmed experimentally by results of Lacy[4])

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4. Lacy, op. cit.

Mixing effects during this period are difficult to estimate, but it appears unlikely that the samples could completely mix in the less than 10 s required for them to spin-down.

GROUND-BASED EXPERIMENTS AND RESULTS

The previous considerations suggested that experimental studies be conducted to determine whether complete mixing can occur in a two-component metal system which is subjected to the described acceleration and temperature profiles. The approach selected was to fabricate samples of the same size and geometry as the flight samples (Fig. 9) but to use two materials which are miscible over their entire range of temperature and composition.

Three material systems were investigated:

In — (In 50 wt% — Bi 50 wt%)

In — (In 70 wt% — Pb 30 wt%)

Eicosane — (Octacosane + black dye).

The samples were encapsulated and mounted on a turntable (Inland, Model No. 712) and subjected to thermal and rotational environments (Fig. 10) as indicated by Table 1. The samples were then cooled to solidification. Any lack of homogeneity in the solidified samples due to incomplete mixing is revealed by density stratification from gravitational settling after spin-down. This density stratification can be observed in the metal samples by slicing them to uniform thickness and subjecting them to X-ray radiography (Fig. 11). Any density differences are revealed due to their effects on the X-ray absorption characteristics of the samples (both density and X-ray absorption increase as the proportion of Pb or Bi in In increases). Density stratification in the paraffin system can be observed visually by the black dye which is added to the denser of the two materials. Results of these experiments are illustrated in Figure 12. This configuration was obtained for all samples. It may be seen that a homogeneous mixture was not achieved.

CONFIGURATION

MORE DENSE MATERIAL A
LESS DENSE MATERIAL B

SAMPLE COMPOSITION

TEST	MATERIAL A	MATERIAL B
1	IN (30 wt%) – Pb	INDIUM
2	IN (30 wt%) – Pb	INDIUM
3	IN (30 wt%) – Pb	INDIUM
4	IN (30 wt%) – Pb	INDIUM
5	IN (50 wt%) – Bi	INDIUM
6	OCTACOSANE + DYE	EICOSANE
7	OCTACOSANE + DYE	EICOSANE
8	OCTACOSANE + DYE	EICOSANE
9	OCTACOSANE + DYE	EICOSANE

Figure 9. Ground-based test sample configuration before melting.
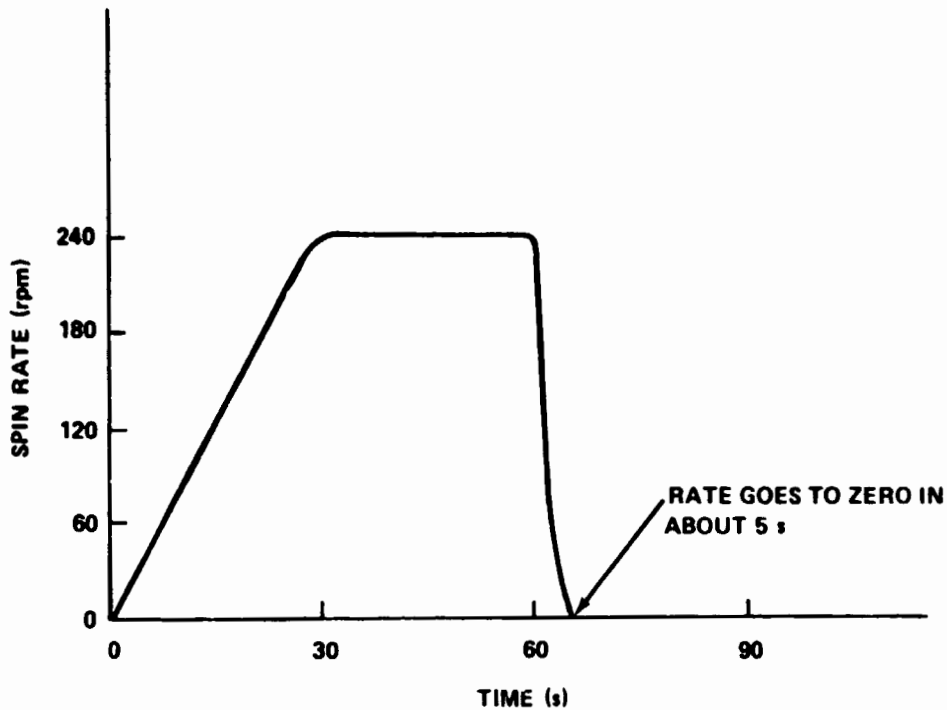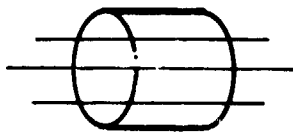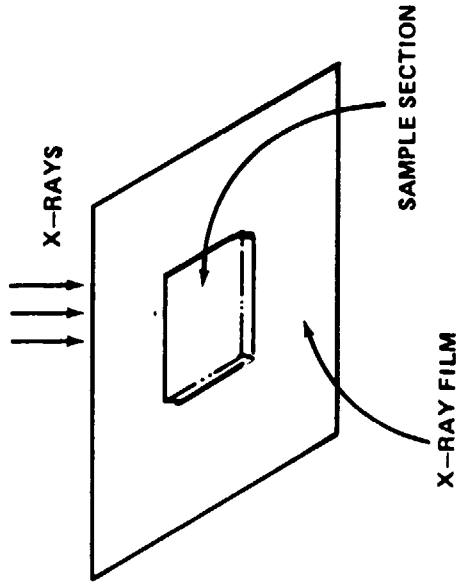
Figure 10.   Laboratory test spin profile.

TABLE 1.   SAMPLE TEST CONDITIONS

Test No.	Sample Composition	Experiment Profile
1	In — (In-Pb)	Heat to 250°C; soak for 1 min; heater off
2	In — (In-Pb)	Heat to 250°C; soak for 20 min; heater off
3	In — (In-Pb)	Heat to 250°C; soak for 15 min; spin-up to 240 rpm; spin-down; soak 90 s; heater off
4	In — (In-Pb)	Same as 3
5	In — (In-Bi)	Same as 3
6	Eicosane-Octacosane	Heat to 125°C; soak for 15 min; spin-up to 240 rpm; spin-down; cool samples
7	Eicosane-Octacosane	Same as 6
8	Eicosane-Octacosane	Same as 6
9	Eicosane-Octacosane	Same as 6

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OPTICAL ANALYSIS OF FILM

LIGHT

FILM WITH
X–RAY IMAGE

TO OPTICAL PROCESSER
AND DIGITAL COMPUTER
IMAGE ANALYSIS

EXPOSE SAMPLE SLICE

X–RAYS

SAMPLE SECTION

X–RAY FILM

SLICE SAMPLE
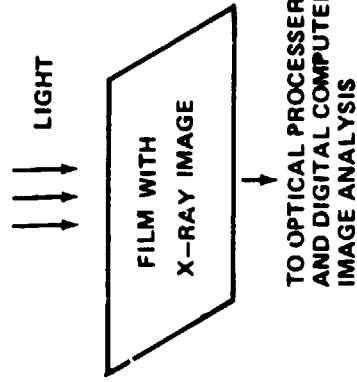
Figure 11. X-ray analysis technique.
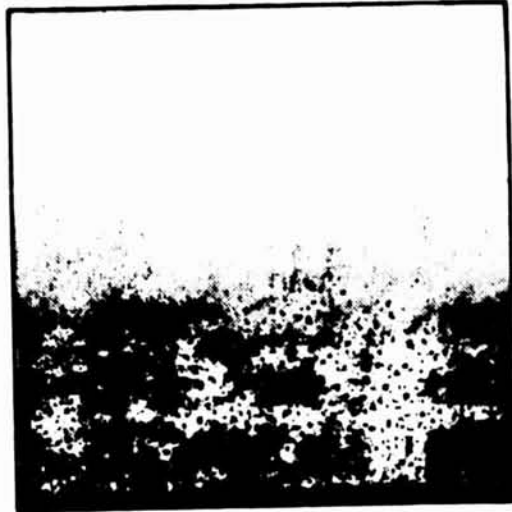
**ALL SAMPLES SHOWED INCOMPLETE
MIXING AS EVIDENCED BY GRADIENTS
OF COLOR OR X-RAY TRANSMISSION
PROPERTIES**

Figure 12. Ground-based test sample configuration
after processing.

SUMMARY AND CONCLUSION

In summary, experiments aboard SPAR II, which were designed to
achieve fine dispersions of In-Al, produced samples in which the In and Al were
segregated on a macroscopic scale. Arguments for kinetic effects during the
sample cool-down phase being responsible for the observed segregation behavior
are unconvincing. Results of studies using miscible systems showed that the
SPAR II heating cycle and spin-up and spin-down were not sufficient to produce
a homogeneous mixture in a two-component system. It can be concluded, there-
fore, that the SPAR II immiscible materials experiments did not achieve homo-
geneity before thermal quench was initiated; consequently, a fine dispersion
would have been impossible to attain. Any future experiments of this type should
include a prelaunch procedure to assure a completely homogeneous sample mate-
rial. This could be most directly achieved by long soak times at a temperature
above the miscibility point of the material system used. These soak times would
be on the order of tens of hours for samples having diffusion coefficients on the
order of 10^{-5} cm^2/s.

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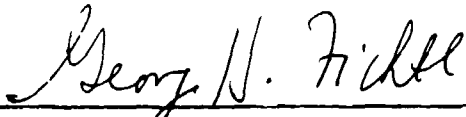   University Press, London, 1968, pp. 124-132.

APPROVAL

MIXING OF TWO LIQUID METALS ON SPAR PAYLOAD DUE TO SPIN-UP AND SPIN-DOWN

By Charles F. Schafer

The information in this report has been reviewed for security classi-fication. Review of any information concerning Department of Defense or Atomic Energy Commission programs has been made by the MSFC Security Classification Officer. This report, in its entirety, has been determ. ed to be unclassified.

This document has also been reviewed and approved for technical accuracy.

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WILLIAM W. VAUGHAN
Chief, Atmospheric Sciences Division

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Director, Space Sciences Laboratory

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