

Particle Engulfment and Pushing by Solidifying Interfaces

Doru M. Stefanescu¹⁾, Frank R. Juretzko¹⁾, Brij K. Dhindaw¹⁾, Subhayu Sen²⁾ and Peter A. Curreli³⁾

1) The University of Alabama, Tuscaloosa, Al; 2) USRA, NASA Marshall Space Flight Center, Huntsville, Al;

3) NASA Marshall Space Flight Center, Huntsville, Al

Abstract

The scientific objectives of the work on Particle Engulfment and Pushing by Solidifying Interfaces (PEP) include: 1) to enhance the fundamental understanding of the physics of interaction between inert particles and the solidification interface, and 2) to investigate aspects of melt processing of particulate metal matrix composites in the unique microgravity environment that will yield some vital information for terrestrial applications. The proposal itself calls for a long-term effort on the Space Station. This paper reports on ground experiments performed to date, as well as on the results obtained from two flight opportunities, the LMS mission (1996) and the USMP-4 mission (1997).

Introduction

During solidification of metal matrix composites the ceramic particles interact with the solidification front. This interaction is responsible for the final microstructure. The solutal and thermal field, as well as fluid motion at the liquid/solid interface influence both interface morphology and the particle/interface interaction itself. It is thus imperative to fully understand the solidification science and transport phenomena aspects associated with the process in order to control it.

The phenomenon of interaction of particles with melt interfaces has been studied since the mid 1960's. While the original interest in the subject was mostly theoretical, researchers soon realized that understanding particle behavior at solidifying interfaces may yield practical benefits. The experimental evidence on transparent organic materials¹ demonstrates that there exist a *critical velocity* of the planar solid/liquid (SL) interface below which particles are *pushed* ahead of the advancing interface, and above which particle *engulfment* occurs.

The main objectives of the experimental and theoretical work were as follows:

- to evaluate the experimental method including sample design, thermal regime, velocity regime, analysis procedures;
- to obtain preliminary data on the critical velocity of PEP in a microgravity environment;
- to interpret these results through mathematical and computational models.

Theoretical

A previously proposed analytical model for PEP was further developed. A major effort to identify and produce data for the surface energy of various interfaces required for calculation was undertaken. The details have been reported⁴. The basic equation for calculation of the critical velocity for the particle-engulfment transition (PET) is:

$$V_{cr} = \left(\frac{\Delta\gamma_o a_o^2}{3\eta K^* R} \right)^{1/2}$$

where $\Delta\gamma_o$ is the surface energy difference between the particle-solid and particle-liquid surface energy, a_o is the atomic or molecular diameter, η is the liquid viscosity, R is the particle radius, and $K^* = K_p/K_L$ is the ratio between the thermal conductivity of the particle (K_p) and of the liquid (K_L).

Experimental

To evaluate the experimental value of PET work was conducted in the following environments:

- ground based
- low-g environment: DC-9
- μ -g environment: LMS, USMP-4

The following systems were used:

- metal/particle: Al /SiC , Al /ZrO₂ , Al-Ni /ZrO₂ , Zn /ZrO₂
- transparent organic/particle: succinonitrile (pure, +0.5% water, +1% water) + polystyrene particles (0.5 to 25 μ m dia.), biphenyl + glass particles (3 to 15 μ m dia.)

Metal/particle systems were investigated under μ -g during the LMS mission, while transparent organic/particle couples were used during the USMP-4 mission.

Metal/Particle Systems and the LMS Mission

Directional solidification ground experiments have been carried out to determine the pushing/engulfment transition for three different metal/particle systems. The matrices were pure aluminum (99.999%), pure zinc (99.95% Zn) and Al – 4.5% Ni alloy. Spherical zirconia particles (500 μ m in diameter) were incorporated in these matrices through mixing in molten state. The particles were non-reactive with the matrices within the temperature range of interest. The experiments were conducted such as to insure a planar solid/liquid interface during solidification. Particle location before and after processing was evaluated by X-ray transmission microscopy for the aluminum-base matrices. All samples were characterized by optical metallography after processing. A clear methodology for the experiment evaluation was developed to unambiguously interpret the occurrence of the PET. A full report on these findings has been published recently².

It was found that the critical velocity for engulfment ranges from 1.9 to 2.4 μ m/s for Al/ZrO₂ and from 1.9 to 2.9 μ m/s for Zn/ZrO₂. No clear PET was found for the Al-Ni/ ZrO₂ system for interface velocities down to 1 μ m/s.

During the LMS Mission three samples were directionally solidified in the AGHF facility, as follows: two pure aluminum (99.999%) 9 mm cylindrical rods, loaded with about 2 vol.% 500 μ m diameter zirconia particles (samples FM1 and FM3); one Al-Ni loaded with the same amount and type of zirconia particles (sample FM2). To validate a cartridge-crucible-sample assembly for the Space Station experiment, two different cartridge designs were used³. For sample FM1 the cartridge included an alumina piston and a graphite spring. The piston-spring system compensated for melting expansion and solidification shrinkage. A simpler design was used for samples FM2 and FM3. It consisted of an expansion reservoir provided at the hot end of the crucible.

The main characteristics of the samples and furnace translation velocity are summarized in **Table 1**. For FM1 a step-wise decreasing regime was used, while for the other two a step-wise increasing regime was chosen. The SL interface velocity resulting from the different furnace translation velocities was calculated based on thermocouple data.

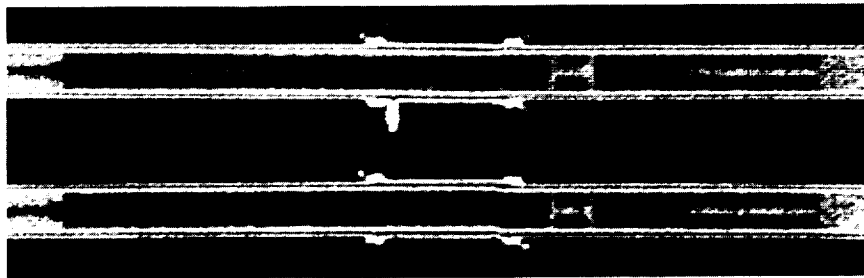
Table 1 Characteristics of flight samples

Flight sample	Material	Ampoule-sample	Velocity regime, μ m/s
FM1	Al - ZrO ₂	spring-piston	20 - 5 - 0.5
FM2	AlNi- ZrO ₂	expansion reservoir	1 - 3 - 9
FM3	Al - ZrO ₂	expansion reservoir	1 - 3 - 9

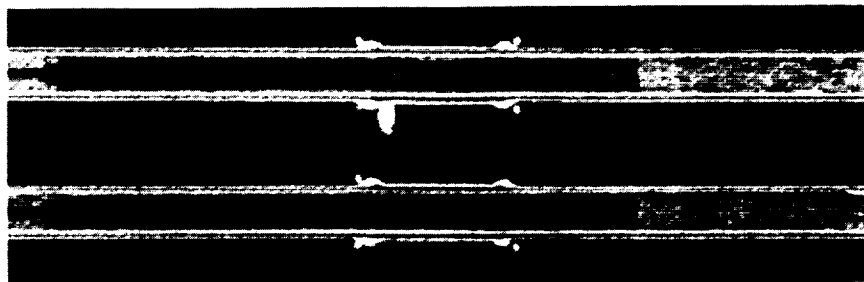
Both the cartridge containing the ampoule, and the ampoule after extraction from the cartridge were examined by X-ray and computer tomography (CT). The results of the cartridge CT evaluation are shown in Fig. 1. The spring-piston assembly used for sample FM1 functioned as

expected. Some liquid Al leak is seen past the fore side of the piston. This leak does not seem to be significant, since no Al is seen in the 90° position of the CT (lower picture on Fig. 1a).

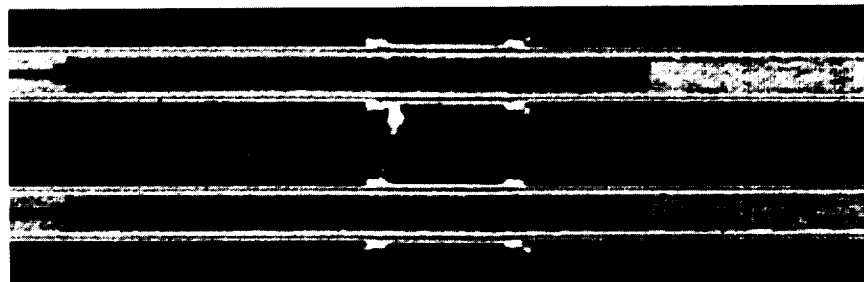
Sample FM2 was less successful. On Fig. 1b it is seen that several voids have formed along the sample. It appears that the liquid metal has fractured in that region resulting in a two-part solid sample: an upper part that is in contact with the alumina plug, and a lower part.



a) sample FM1



b) sample FM2



c) sample FM3

Fig. 1 CT images of the flight cartridge - ampoule assembly³.

Sample FM3 behaved as expected. On Fig. 1c it is seen that the shrinkage cavity was positioned between the metal and the alumina plug. The sample itself appears to have no significant shrinkage porosity or voids. This was also confirmed through metallographic examination.

For the Al/ZrO₂ sample it was found that a PET occurred in the velocity range of 0.5 to 1 μm/s (see Fig. 2 and Fig. 3). This is smaller than the ground PET velocity of 1.9 to 2.4 μm/s. It demonstrates that natural convection increases the critical velocity. A detailed report of these results has also been published⁴. For the Al-Ni/ ZrO₂ system no PET was found down to the lowest velocity used during the experiment which was of 1 μm/s.

A comparison between measured and calculated critical velocity for the metallic systems studied on ground and μg are given in Table 2. It is seen that the predicted value lies within the experimentally evaluated μg value.

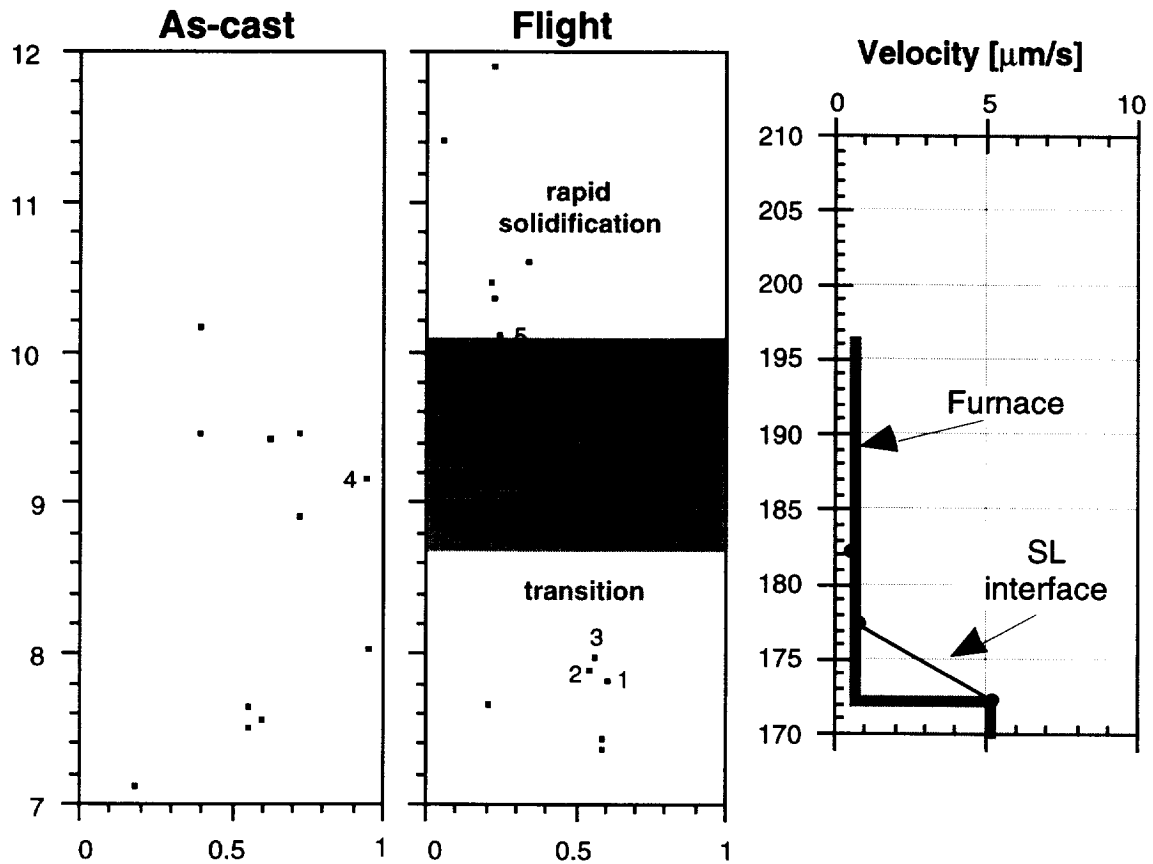


Fig. 2 Summary of XTM evaluation of particles positions in sample FM1 in the region of furnace translation rate of $0.5 \mu\text{m/s}$ ⁴

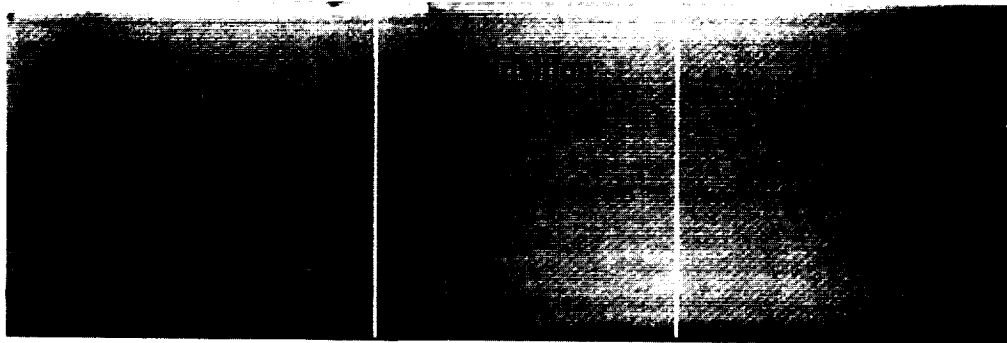


Fig. 3 XTM image of flight sample FM1. Particles are engulfed at $5 \mu\text{m/s}$ and in the transient region. No valid engulfed particles are present in the $0.5 \mu\text{m/s}$ region ⁴

Table 2 Experimental and theoretical values for the critical PET velocity

Data from	Critical Velocity, $\mu\text{m/s}$	
	Al/ZrO ₂	Zn/ZrO ₂
Ground experiments	1.9 – 2.4	1.9 – 2.9
LMS	0.5 – 1.0	-
Model	0.775	0.672

Organic Material/Particle Systems and the USMP-4 Mission

Ground and parabolic flight experiments were also performed with a biphenyl matrix / spherical glass particles, and succinonitrile matrix / polystyrene particles. Two experimental setups were used: a horizontal gradient heating facility (HGF) for horizontal solidification, and a Bridgman-type furnace (BF) for vertical solidification. The convection level during solidification in the HGF was varied by changing the distance between the glass slides containing the composite sample. The BF was used on ground and during parabolic flights, and thus the convection level was changed by alternating low-gravity and high-gravity solidified regions. It was found that the convection level and/or particle buoyancy significantly influences the critical velocity for particle engulfment. At higher natural convection during solidification the critical velocity increases by up to 40%. At very high convection levels engulfment may become impossible because particles fail to interact with the interface. A detailed account of these experiments was previously reported⁵.

In the vertical configuration Stokes settling velocity appeared to have an influence on the critical velocity for particle engulfment. This was more pronounced for thicker sample cells since the settling velocity is expected to increase with increase in sample cell thickness. The effect of Stokes velocity was to decrease the critical velocity by up to 40%. This is also illustrated by the slight increase in critical velocity when experiments were performed at lower gravity levels.

During the USMP-4 mission similar experiments on biphenyl matrix and spherical glass particles, and succinonitrile matrix with polystyrene particles were performed. The standard procedure used for the SCN/polystyrene samples consisted in directional solidification (DS) of the sample through incremental velocity changes (increase or decrease). If agglomerates or too many particles accumulated at the interface during pushing, rapid DS (10 or 15 $\mu\text{m/s}$ for 30 to 50 s) was used to engulf the particles and clear the interface. Then, incremental velocity solidification was resumed.

Flight and ground experimental results together with theoretical predictions are summarized in Fig. 4. Triangles describe engulfed particles, while circles pushed particle. It is seen that a lower and upper limit for PET exists. Between these two limits, some particles are pushed and some are engulfed. It is also seen that the ground critical velocity lies significantly above the one found in μg , as expected.

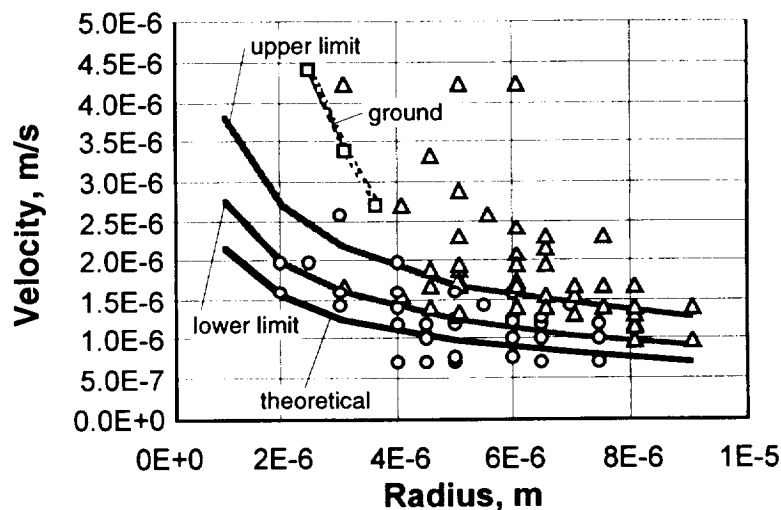


Fig. 4 USMP-4 results for SCN/polystyrene.

The theoretical critical velocity is slightly lower than the lower experimental limit. The molecular diameter calculated from the molar volume of SCN ($0.081 \text{ m}^3/\text{kmol}$) was $a_0 = 1.14 \cdot 10^{-9} \text{ m}$.

Evaluating individual particles was more difficult for the biphenyl samples because biphenyl became opaque after resolidification. Some preliminary results are given in Fig. 5.

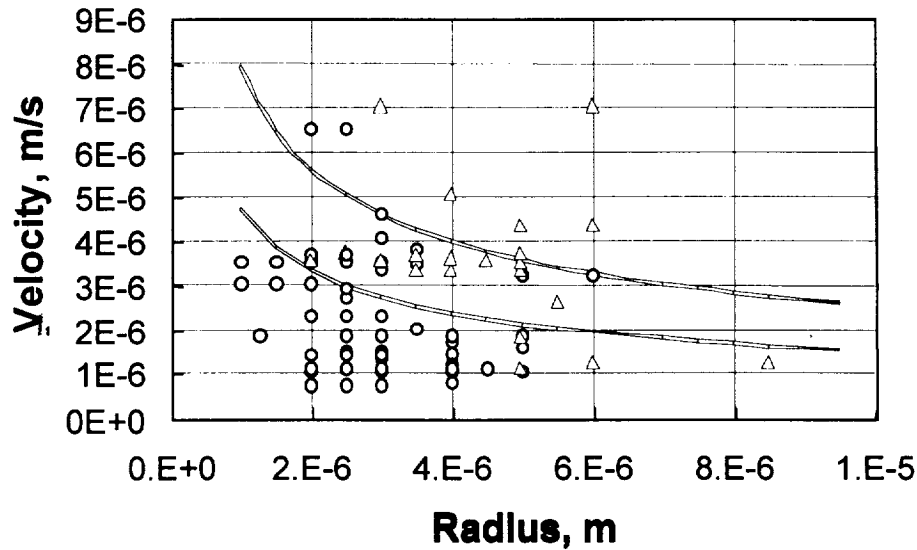


Fig. 5 USMP-4 results for biphenyl/glass.

Another procedure consisted in DS at a set velocity, after which rapid DS was used to engulf the particles. This was followed by rapid directional remelting which left behind in the liquid a band of particles (that were pushed at the set velocity). Since the liquid is transparent, it was possible to measure the particle size. In developing this method full advantage was taken of the real-time video down-link and direct interaction with the astronauts. Measurements with this procedure, as well as theoretical calculation are not available yet.

The experiments clearly demonstrated that the particles produced interface instability. For example, in one case, the interface became unstable at $2.8 \mu\text{m/s}$ when a large amount of particles were pushed ahead of the interface. However, after the interface was "cleaned" by running at $10 \mu\text{m/s}$, the interface did not break down until the velocity reached $5.2 \mu\text{m/s}$.

Pushing of large agglomerates of particles was systematically observed. On ground agglomerates are broken down by the convective flow and/or easily engulfed.

Additional observations were made on shrinkage flow into the interface, and on the effect of g-jitters on particle behavior at the interface. It was noticed that every time the verniers were fired particles and agglomerates were jerked along the liquid/solid interface.

References

- ¹ S.N. Omenyi and A.W. Neumann: *J. Appl. Phys.*, 1976, vol.47, no.9, pp. 3956-62
- ² F. R. Juretzko, B. K. Dhindaw, D. M. Stefanescu, S. Sen and P.A. Curreri, *Metall. and Materials Trans.*, **29A** (1998) 1691
- ³ D. M. Stefanescu, B. K. Dhindaw, F. R. Juretzko, P.A. Curreri and S. Sen, in *Life and Microgravity Spacelab (LMS) Final Report*, J. P. Downey compiler, Marshall Space Flight Center, Alabama (1998) 76
- ⁴ D. M. Stefanescu, F. R. Juretzko, B. K. Dhindaw, S. Sen and P.A. Curreri, *Metall. and Materials Trans.*, **29A** (1998) 1697
- ⁵ S. Sen, B.K. Dhindaw, D.M. Stefanescu, A. Catalina and P.A. Curreri, *J. Crystal Growth*, **173** (1997) 574