Particle Engulfment and Pushing by Solidifying Interfaces

Doru M. Stefanescu[1], Frank R. Juretzko[1], Brij K. Dhindaw[1], Subhayu Sen[2], and Peter A. Curreri[3]

[1] University of Alabama, Tuscaloosa, Alabama
[2] Universities Space Research Association, NASA Marshall Space Flight Center, Alabama
[3] NASA Marshall Space Flight Center, Huntsville, Alabama

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 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 Life and Microgravity Spacelab (LMS) mission (1996) and the Fourth United States Microgravity Payload (USMP-4) mission (1997). The main objectives 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; and,
- to interpret these results through mathematical and computational models.

During these experiments, metal/particle and transparent organic/particle couples were used. Directional solidification ground experiments have been carried out to determine the pushing/engulfment transition (PET) for two different metal/particle systems. The systems chosen were aluminum/zirconia particles and zinc/zirconia particles. Pure metals (99.999% Al and 99.95% Zn) and spherical particles (500 µm in diameter) were used. 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 Al/ZrO_2 samples. 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 pushing-engulfment transition. It was found that the critical velocity for engulfment ranges from 1.9 to 2.4 µm/s for Al/ZrO_2 and from 1.9 to 2.9 µm/s for Zn/ZrO_2.

Results of the directional solidification experiments on PEP conducted on the Space Shuttle *Columbia* during the LMS mission are also reported. Two pure aluminum (99.999%) 9 mm cylindrical rods, loaded with about 2 vol.% 500 µm diameter zirconia particles were melted and resolidified in the microgravity (µg) environment of the Shuttle. One sample was processed at step-wise increased solidification velocity, while the other at step-wise decreased velocity. It was found that a PET occurred in the velocity range of 0.5 to 1 µm/s. This is smaller than the ground *PET* velocity of 1.9 to 2.4 µm/s. This demonstrates that natural convection increases the critical velocity.

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 predicted *PET* critical velocity for Al/ZrO_2 was of 0.775 µm/s.

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.

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 mm/s for 30 to 50 s) was used to engulf the particles and clear the interface. Then, incremental velocity solidification was resumed.

Because biphenyl became opaque after resolidification, a different procedure was used. It 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.

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