MATERIALS SCIENCE EXPERIMENTS UNDER MICROGRAVITY - A REVIEW OF HISTORY, FACILITIES, AND FUTURE OPPORTUNITIES

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Abstract

Materials science experiments have been a key issue already since the early days of research under microgravity conditions. A microgravity environment facilitates processing of metallic and semiconductor melts without buoyancy driven convection and sedimentation.
Hence, crystal growth of semiconductors, solidification of metallic alloys, and the measurement of thermo-physical parameters are the major applications in the field of materials science making use of these dedicated conditions in space. In the last three decades a large number of successful experiments have been performed, mainly in international collaborations. In parallel, the development of high-performance research facilities and the technological upgrade of diagnostic and stimuli elements have also contributed to providing optimum conditions to perform such experiments. A review of the history of materials science experiments in space focussing on the development of research facilities is given. Furthermore, current opportunities to perform such experiments onboard ISS are described and potential future options are outlined.

Introduction

A microgravity environment facilitates processing of fluids without buoyancy driven convection and sedimentation. Ideally, the absence of gravity-driven buoyancy convection features a purely diffusive transport; the absence of sedimentation leads to homogeneous particle distribution in mixtures. For materials science these effects are mainly relevant for experiments aiming at crystallisation of bulk materials from the fluid state (melt or vapour phase) and on the determination of thermo-physical parameters in the under-cooled regime.

Materials science experiments have been therefore a prominent topic, already since the early days of research under microgravity (µg) conditions. The first experiments under reduced gravity were reported during the fly-back phase of the Apollo 14 moon mission in 1971 [1]. A couple of demonstration experiments have been performed by the astronauts A. Shephard, S. Roosa, and E. Mitchell to experimentally assess the advantage of materials processing in space. Composite casting has been selected as one reference experiment for materials science. 11 different samples were melted and re-solidified in order to permit evaluation of the particle re-distribution. For example, it could be shown that a mixture out of paraffin, Be-Cu particles and Ar gas displayed a somewhat homogeneous distribution after re-solidification under µg conditions. Fig. 1 shows a photograph of the mixture after cool-down and the simple setup to perform this experiment.
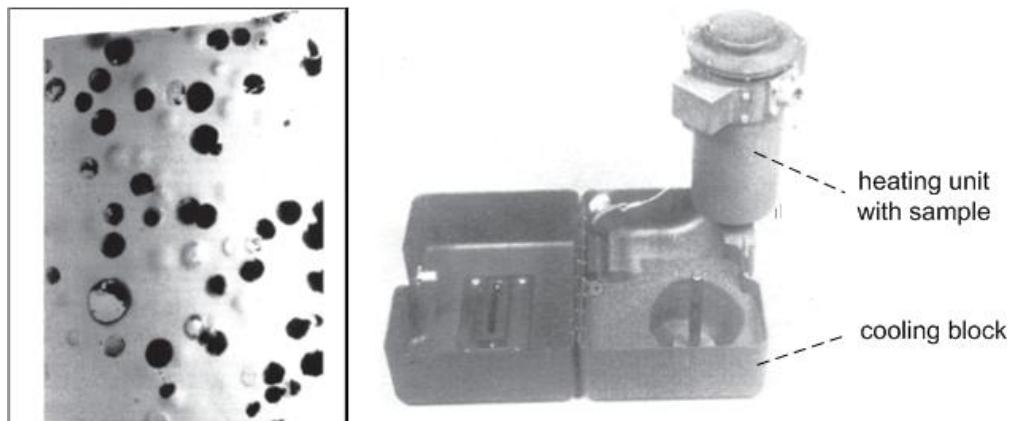

Figure 1.   Composite casting experiment on Apollo 14. Left part: Re-solidified mixture out of paraffin, Be-Cu particles, and Ar gas bubbles; right part: Experimental apparatus

These experiments represented a pioneering work in terms of a first proof-of-principle and hence opened the door for a long series on investigations under space conditions on various carriers and missions, starting with Skylab, the Apollo/Soyuz mission, Spacelab, other shuttle experiments, sounding rockets, free flyers, and finally on space stations like Mir or ISS.

In this paper a review on the objectives, performance and results of high-lighted experiments in the main fields for materials science research is presented
- Crystal growth
- Solidification
- Diffusion processes
- Measurement of thermo-physical properties.

Furthermore key technology improvements which have lead to innovative scientific research are reported and finally an overview on currently available facilities, opportunities, and recognizable trends is given.

Materials Science Experiments

During the last three decades the general objectives that materials science experiment in space have thus been focussed
- To understand fundamental mechanism and processes
- To validate numerical models
- To provide benchmark data for industrially important materials
- To measure accurately thermo-physical properties which can not easily reached on earth.

In the following sections the different areas of materials science are reviewed and the results of high-lighted experiments are described.

Crystal growth

Crystal growth of single-crystalline materials from the liquid and gas phase as well is performed very close to the thermodynamic equilibrium, i. e., with very low growth rates and having a flat or only smoothly curved solid/liquid interface. Hence, these processes are rather sensitive to small perturbations of all kinds, maybe caused by short-term fluctuations in the temperature field of the furnace, variations in the ganging of the mechanical drive of the heater or sample, or by different types of time-dependent convective flows in the melt [2]. The first two issues must be addressed by the design and performance of the facility, processing under μg conditions could help to reduce the latter one.
In the early days the main target for crystal growth experiments in space - especially for semiconductors - was the improvement of material quality caused by the absence of buoyancy effects. The first experimental results however, were quite disillusioning: A. Eyer et al. discovered during the float-zone growth of Si that Marangoni convection is the dominant factor in the melt flow, more than outbalancing the absence of buoyancy-driven convective motions [3]. In a comprehensive survey K. W. Benz has described the consequences drawn by the community from Eyer's early experiment and the resulting metamorphosis of objectives for semiconductor crystal growth in space [4]. The most striking results for performing crystal growth experiments under reduced gravity conditions which have been obtained so far are summarised below:

- To attain larger diameter for float-zone crystals [5]
- To measure Marangoni numbers without overlaying buoyancy flows [6, 7]
- To study the influence of soluto-capillary flow [8, 9]

– To study the influence of fluid flow stimuli like magnetic fields or mechanical vibrations [10, 11, 12]
– To allow for numerical calculations under well-defined conditions [13]
– To study detached solidification [14]

As example the float zone growth of Ga-Sb [5] which yielded a significant reduction of striations in the space grown crystal compared to its earth-grown counterpart, is illustrated in Fig. 2.

These investigations were also fostered by a steady improvement in the technical capabilities of the respective facilities. For example, mirror furnaces were the working horses in the early days on S/L, sounding rocket, and EURECA missions. They were robust, easy to operate, and allow for a direct optical control of the solid/liquid [15]. But the thermal profile was fixed and the temperature gradient at the S/L IF could not be adjusted. Nowadays multi-zone furnaces built up by several individual heaters with a high-stability temperature control system are available onboard ISS. With these furnaces the thermal gradient at the phase boundary can be adjusted over a wide range, and perform various types of experiments can be performed [16, 17]. Furthermore, in order to suppress stochastic flows which are caused by residual accelerations, the implementation of magnet systems inducing a well-defined flow in the melt into these new facilities has been pursued in the last decade. First experiments performed on sounding rocket missions revealed indeed that the melt flow can be substantially affected by a rotating magnetic field [18] in microgravity, thus providing ideal reference cases for testing and improving numerical models on the melt flow.
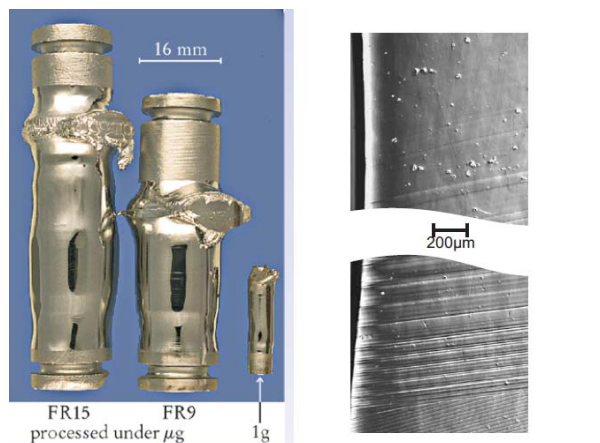

Figure 2.   Crystal growth of GaSb on Spacehab-4 mission. Left part: Size comparison between earth grown and μg crystals. The right part illustrates the significantly reduced micro-segregation under μg (less striations in upper photograph)

Actually, two major experiments from field of the crystal growth are envisaged for the ISS: RDGS and SISSY. The first one is targeted to determine the effects of "detached" Bridgman growth in comparison to regular Bridgman growth and float-zone (FZ) growth on the defect formation and distribution in Ge-Si crystals [19]. The international cooperation SISSI aims at

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investigating the gravitational effects on heat and mass transport phenomena in directional solidification of upgraded metallurgical silicon for photovoltaic applications [20].

Solidification

One key aspect of the solidification that influences the microstructure is the shape of the solid-liquid interface in a solidifying material. At low growth rates the interface is planar like for semi-conductors. Industrially relevant metallic materials crystallise at much higher growth rates. As the rate of growth increases, the interface develops a corrugated texture until three dimensional cells form in the solid. Even higher growth rates cause the formation of dendrites. The development of these different interface shapes and the transition from one shape to another is controlled by the morphological stability of the interface. Gravity represents one of the most influencing factors to this stability. In particular, buoyancy-driven convection can influence the stability and, thus, the shape of the solidifying interface. Hence, processing under reduced gravity imposes therefore well-defined conditions for crystallisation. Thus, the results obtained so far from microgravity experiments about the conditions under which certain types of solidification boundaries appear could help to explain the formation of the crystalline structure of a material, to validate numerical models, and to improve terrestrial applications [21].
In the recent decades the subsequently listed most striking results have been obtained in microgravity experiments:

- Study of morphological stability in immiscible systems [22]
- Processing of eutectics [23]
- Melt interface stability studied by Seebeck coefficient variation [24]
- Dispersed systems [25]

In addition to the mentioned issues dendritic growth is the common mode of solidification applied for metals and alloys under low thermal gradients. The growth of dendrites in pure melts depends on the transport of latent heat from the moving crystal–melt interface and the influence of weaker effects like the interfacial energy. Experimental data for critical tests of dendritic growth theories remained limited because dendritic growth can be complicated by convection. A fundamental understanding of dendritic solidification is necessary to correct mathematical models that will provide a basis for improved industrial production techniques. Prof. M. Glicksman and his research team have performed a series of pioneering and quite successful experiments on the mechanisms of dendrite formation by eliminating buoyancy-induced convection in the Isothermal Dendritic Growth Experiment (IDGE). This facility has been flown three times as part of the United States Microgravity Payload (USMP) series. During the first two flights of IDGE on the shuttle, the IDGE flight hardware grew and photographed individual dendrites of the material succino-nitrile (SCN) as they solidified at various temperatures (see Fig. 3). The third flight of IDGE on USMP–4 has used a different sample material, pivalic acid Dendrite growth velocities could be measured and the researchers were able to compare the results with theoretical predictions and to answer questions on fundamental characteristics on dendrite growth [26, 27, 28].
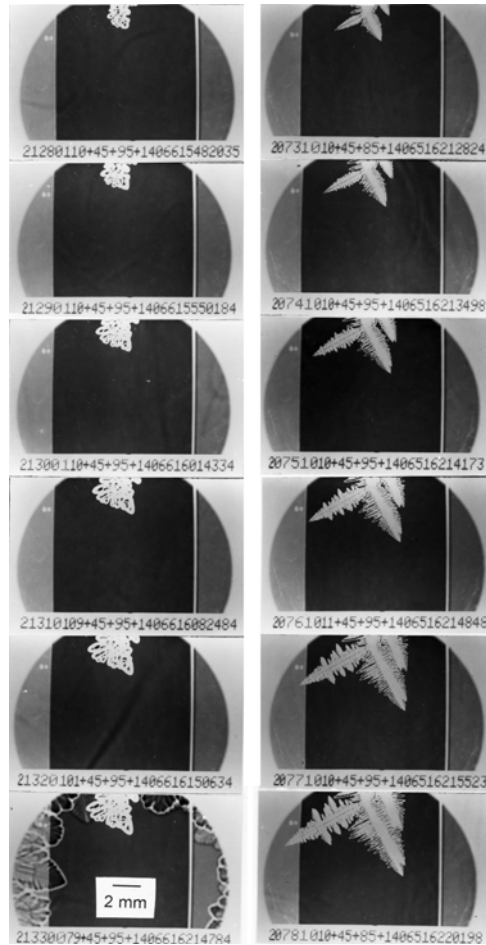
Figure 3. Example of two image sequences of dendritic growth for SCN at two supercoolings: Left: 0.1 K (photographs taken every 700 s); right: 1 K (photos every 6.75 s)

The modular experimental facility MSL (Materials Science Laboratory) [16] which is implemented in the US-module destiny of the ISS is successfully being operated since 2009. It enables in its current condition to perform investigations on

- Columnar to Equiaxed Transition in Solidification Processing (CETSOL) [29]

- Microstructure Formation in Casting of Technical Alloys under Diffusive and Magnetically Controlled Convective Conditions (MICAST) [30]

- Solidification Along A Eutectic Path in Ternary Alloys (SETA) [31]

In the near future it is considered to develop an MSL insert for in-situ diagnostic of dendritic growth by radiography [32].

Diffusion

The determination of diffusion constants in liquids allows for validating theoretical models on diffusive transport phenomena at one side. Furthermore, if a purely diffusion-controlled flow regime is established under μg conditions the exact knowledge of the diffusion constants is essential for modelling the fluid flow in the melt during crystal growth and solidification processes.

Under terrestrial conditions the measurement of diffusion constants in the liquid state is always overlaid by gravity-driven convective flows. For this reason the measurement of diffusion constants was already one of the major topics for performing experiments under microgravity conditions in the early years. The first measurements of diffusion constants were performed during S/L missions or on the MIR station using the long-capillary (LC) technique [33-35]. For single-component systems the experiments indeed disclosed much lower values for the diffusion constants than obtained in earth-bound experiments [33]. This technique, however, leads for multi-component systems to segregation effects resulting in an additional mass transport which give rise to a systematic measurement error. This drawback can be overcome by a stable density layering of the sample materials. Thereby the density has to increase parallel to the gravity vector resulting in a density gradient corresponding to the concentration gradient.

By the implementation of the shear-cell technique other drawbacks (free surface convection, diffusion during heat-up and cool-down) of the LC technique can be avoided: A rapid partitioning the sample material into several small volumes after processing but before cool-down by means of a dedicated mechanical mechanism leads to a quasi-frozen concentration distribution for off-line analysis. On the un-manned FOTON-12 mission pioneering measurements of diffusion constants employing the shear-cell technique up to a temperature of 1200°C have been successfully carried out [36, 37]. The diffusion constants of several multi-component systems have been successfully measured on the FOTON-M2 mission in 2005 [38].

A further innovation has moved in the field during the recent years, the in-situ determination of diffusion profiles via radiography. DLR has initiated the development of the DIXI heater insert for the MSL facility onboard ISS. This facility would allow performing in-situ diagnostics for high-temperature diffusion experiments as illustrated in Fig. 4 [32].
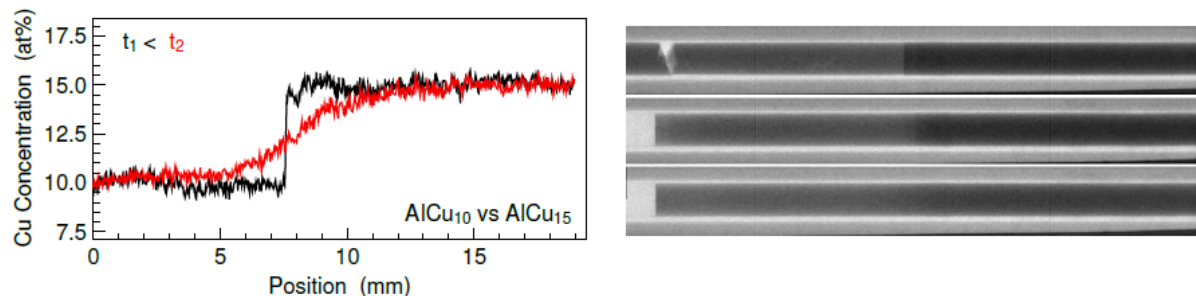

Figure 4.   The concentration profiles of a Al-Cu alloys can be deduced from the grey values of an in-situ radiographic image (right) in the DIXI facility on ground [32]

The measurement of diffusion constants of self-diffusion in elements like Si and of multi-component systems where no stable density layering can be establish (Si-Ge) represents the actual trends for future experiments. Today, all the envisaged experiments on the ISS from European- of Japanese-coordinated research groups favour the combination of the shear-cell technique with in-situ radiography.

Levitation techniques

Experiments on crystal growth and solidification are generally bound to a crucible containing the sample material. The crucible is integrated into a cartridge if a closed containment to the process environment has to be provided. This design affects the measurement of surface-sensitive properties like viscosity and surface tension and prevents to reach significant undercooling of the liquid sample as well. Furthermore, the chemical compatibility between the crucible and sample material at elevated temperatures imposes often severe problems leading in the worst case to a complete exclusion from processing for rather critical metals than Ti where no appropriate high-temperature resistant crucible materials is available.

To overcome these obstacles levitation methods have been established into in materials science research facilities. Two main techniques have been successfully applied in the past:
- Electrostatic levitation
- Electromagnetic levitation

In electrostatic levitators the sample is electrically charged, levitated in a static electric field, and heated by a laser. This enables to process preferably non-metallic elements and oxide materials. Both, JAXA and NASA are fostering the development and operation of an *Electrostatic Levitator (ESL)* on ground for a potential later implementation on the ISS [39, 40].

Electromagnetic levitation allows processing of electrically conductive samples including pure metals, alloys and semiconductors. The sample is thereby placed in the center of a coil system which is part of an oscillating RF circuit and generates an RF electromagnetic field. The interaction of eddy currents induced in the sample with the electromagnetic field leads to a positioning of the sample at the center of the coil system. Heating is achieved by ohmic losses of the eddy currents flowing in the sample [41, 42].

For ground-based experiment strong levitation fields are needed for both techniques in order to counteract gravity. This leads to a number of drawbacks like large power consumption, difficult control of sample positioning, convective flows in the melt, and a deformed geometrical sample shape. These drawbacks can be eliminated if levitation is performed under microgravity conditions where only small levitation forces are required to compensate for the residual accelerations. Moreover, convective flows are significantly reduced and the sample shape is nearly spherical.

Electromagnetic levitation experiments in space have already a long-lasting successful history: The DLR facility TEMPUS (see Fig. 5) was operated on three Spacelab missions (IML-2 in 1994, MSL-1 and MSL-1R in 1997) and provided a wealth of scientific data [41, 43].
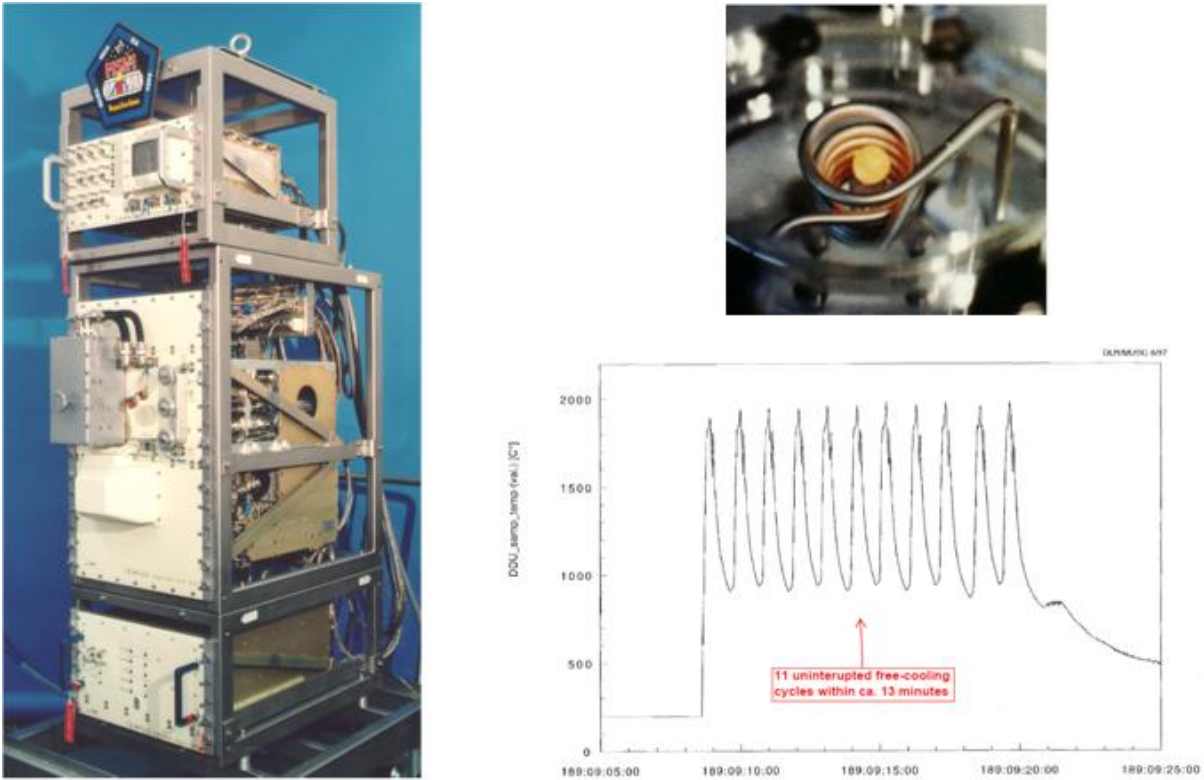
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Figure 5: TEMPUS facility (left). In the upper right part the levitated molten sample is shown, the lower right part displays the undercooling of a levitated Zr sample as measured during the MSL-1 (see [44]).

One of the prominent scientific topics of these missions was the investigation on the kinetics of solid nucleation in Zr. Bayuzick and co-workers could perform 122 cycles of undercooling with a ΔT=333°C below the nominal solidification temperature and could study the influence of fluid flow regimes on the nucleation kinetics via variation of the cooling rate [44].

Compared to crystal growth or solidification experiments the duration of a levitation run is short and the sample can be re-processed several times enabling to perform systematic investigations even on one single mission. For the purpose of extending the experimental capabilities even further, currently the ISS version of this facility the *Electromagnetic Levitator (EML)* is being developed and will be operational in space from 2014 [42].

In addition to the performed Spacelab experiments a number of campaigns have been conducted on sounding rockets and on parabolic flights as well in the recent decades. This makes electromagnetic levitation the most prevalent technique for materials science investigations in space.

Outlook

Microgravity has to be regarded as tool which can be employed to provide dedicated experimental conditions like the application of external fields or forces does. In all relevant research areas of materials science it could be shown that a microgravity environment is advantageous for the investigation of specific subjects. Numerous distinguished experiments have been performed and were well received within the science community. The character of these results, however, can be portrayed in most cases as proof-of-principle or benchmarks. Due to the lack of continuous experiment opportunities so far and also due to the long duration of experiments mainly single-shot experiments have been conducted - experiments using electromagnetic levitation being an exception here because of the reasons mentioned above.

The next coherent step in this context would be to convert the nature of material science research from primarily benchmarking to a more systematic research. Utilization of the ISS offers exactly this perspective and gives the unique possibility to initiate and perform intense research programs embedded in international frameworks especially if emphasizing the extended utilization period up to 2020. The facilities to perform the above described class of experiments are either already implemented (MSL, GHF, DECLIC), in preparation (EML), planned (ESL, TRANSPARENT ALLOYS), or addressed in several outstanding science proposals and consequently envisaged by the Agencies. The extended capabilities leave certainly much room for an extension of the science community resulting in a higher science output.

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