

Studies of Thermophysical Properties of Metals and Semiconductors by Containerless Processing under Microgravity

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Abstract

Electromagnetic levitation under microgravity provides unique opportunities for the investigation of liquid metals, alloys and semiconductors, both above and below their melting temperatures, with minimized disturbances of the sample under investigation. The opportunity to perform such experiments will soon be available on the ISS with the EML payload which is currently being integrated. With its high-performance diagnostics systems EML allows to measure various physical properties such as heat capacity, enthalpy of fusion, viscosity, surface tension, thermal expansion coefficient, and electrical conductivity. In studies of nucleation and solidification phenomena the nucleation kinetics, phase selection, and solidification velocity can be determined. Advanced measurement capabilities currently being studied include the measurement and control of the residual oxygen content of the process atmosphere and a complementary inductive technique to measure thermophysical properties.

Introduction

Electromagnetic levitation provides unique opportunities for the investigation of electrically conductive liquid metals, alloys and semiconductors (if pre-heated or doped), both above and below their melting temperatures. In particular, the undercooled regime where the sample is liquid at temperatures below its equilibrium melting temperature is accessible through electromagnetic levitation thus allowing the determination of thermo-physical properties of materials in this particular state as well as studying of nucleation and solidification phenomena. Thermo-physical properties that can be measured comprise heat capacity, enthalpy of fusion, thermal transport coefficients in the liquid phase, viscosity, surface tension, thermal expansion coefficient, hemispherical emissivity, fraction solid and electrical conductivity. In studies of nucleation and solidification phenomena the nucleation kinetics and solidification velocity can be determined. [1], [2]

Ingredients

In order to prepare the undercooled state and to perform experiments aiming at the determination of the thermo-physical properties without adverse impacts related to chemical reactions, heterogeneous nucleation, or lack of visibility, the following ingredients are essential:

2.1. Electromagnetic levitation

Electromagnetic levitation allows processing of the samples without contact to a container. A wide range of electrically conductive samples including pure metals, alloys and semiconductors can be processed with this technique. The sample is 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 displacement force which keeps the sample at the center of the coil system. Heating is achieved by ohmic losses of the eddy currents flowing in the sample. In ground-based experiments the required levitation force and therefore electromagnetic field strength to counteract gravity is so large that many materials are melted just by applying the field to position them. Heating and positioning of the

sample are therefore not independent and the undercooled regime of samples with low melting temperatures is not accessible. Furthermore, the electromagnetic pressure exerted by the strong fields leads to strong convection and a deformation of the liquid samples which are not compatible with many experiment objectives such as the determination of viscosity and surface tension. The need for lower electromagnetic field strengths thus becomes evident which means that gravity has to be largely eliminated in order to perform experiments with undisturbed samples over a wide temperature range [4-8].

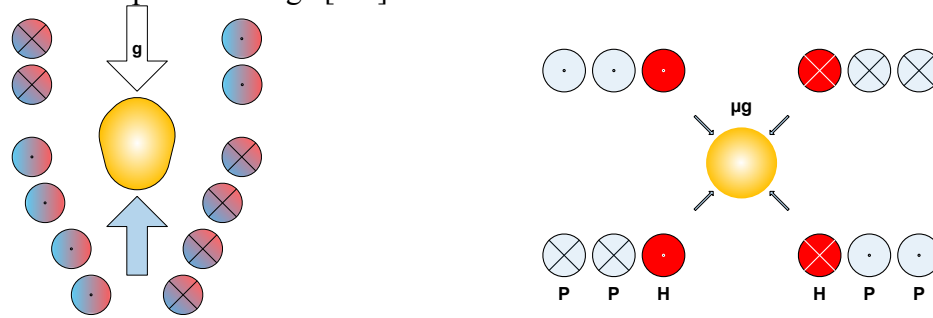


Figure 1. Principle design of levitation coils for levitation under 1g (left) and μg (right). In a 1g levitation coil the positioning force has to counteract gravity, therefore strong electromagnetic fields are needed just for the positioning of the sample, which leads to a strong heating and deformation of the sample (left). In contrast, the forces required to position a sample in a μg environment are orders of magnitude smaller, therefore the heating and deformation of the sample due the positioning field are negligible. As a consequence, heating and positioning of the sample can be controlled largely independently, e.g. by using dedicated coils for heating and positioning, respectively (right). Note that this illustrative sketch does not show the current EML flight coil geometry.

2.2. Ultraclean process environment:

In order to reduce the potential for heterogeneous nucleation as much as possible an ultra-high vacuum or ultra-pure noble gas process atmosphere is necessary. In particular, the amounts of reactive trace gases, like for example oxygen, need to be minimized.

2.3. Contactless diagnostics

The diagnostic instruments have to be compatible with the containerless processing principle as well, such as pyrometers for measurement of the sample temperature and various dedicated video cameras for specific purposes, e.g. cameras with high spatial or temporal resolution.

2.4. Sample handling

Even though the samples are processed without contact to a container wall they need to be carefully prepared and handled during the experiment. Typical experimental set-ups provide the possibility to store several samples, to select a dedicated sample for processing and to transfer the selected sample into the coil. Therefore, the samples are placed in so-called sample holders made of chemically inert thermal shock resistant material which are connected to a transfer mechanism and provide enough free space for the sample such that the containerless principle is respected during processing.

3. Undercooling at Work

Driven by the size of the RF coils which are optimized for positioning and heating efficiency under microgravity the samples are spherical with diameters typically ranging from 5 to 8 mm. The process environment for the samples is either UHV or ultra pure noble gas (He or Ar) up to 400 mbar.

A typical experiment consists of several melt cycles. Starting from the solid state the sample is heated up above the melting temperature (overheating of up to 300 K), and then the heater power into the sample is reduced. During the subsequent cooling phase, the majority of the experimental data are gathered. Depending on its properties the sample undercools to temperatures below the melting temperature until solidification sets in rapidly and the heat of fusion is released in a very short time such that the sample temperature rises again to the melting temperature, a phenomenon known as recalescence. During the following cool down of the solid sample the cooling rate can be enhanced by additional convective cooling with the process gas being circulated through a pipe and filter system and released through a nozzle at the sample's vicinity. A typical melt cycle is shown in figure 2.

Due to the small size and related low heat content of the sample the typical duration of a melt cycle is in the order of seconds to minutes, depending on the sample material and the definition of the process, such that many such melt cycles can be performed during a the time frame available for an on-orbit experiment which is in the order of hours.

At high temperatures samples processed in ultra-high vacuum may evaporate some material which leads to a deposition of evaporated material on the inner surfaces of the experiment set-up in a physical vapor deposition process. Components and assemblies in the vacuum chamber that are sensitive to deposition of evaporated sample material like the components of the optical path from the sample to the pyrometer and video cameras have to be protected from deposition by e.g. exchangeable double mirror systems. If samples are processed in a noble gas atmosphere the evaporated material does coagulate into microscopic dust particles which are toxic and need to be removed from the experiment setup by a dedicated gas cleaning system.

After processing samples can be retrieved from the facility and returned to ground for further evaluation.

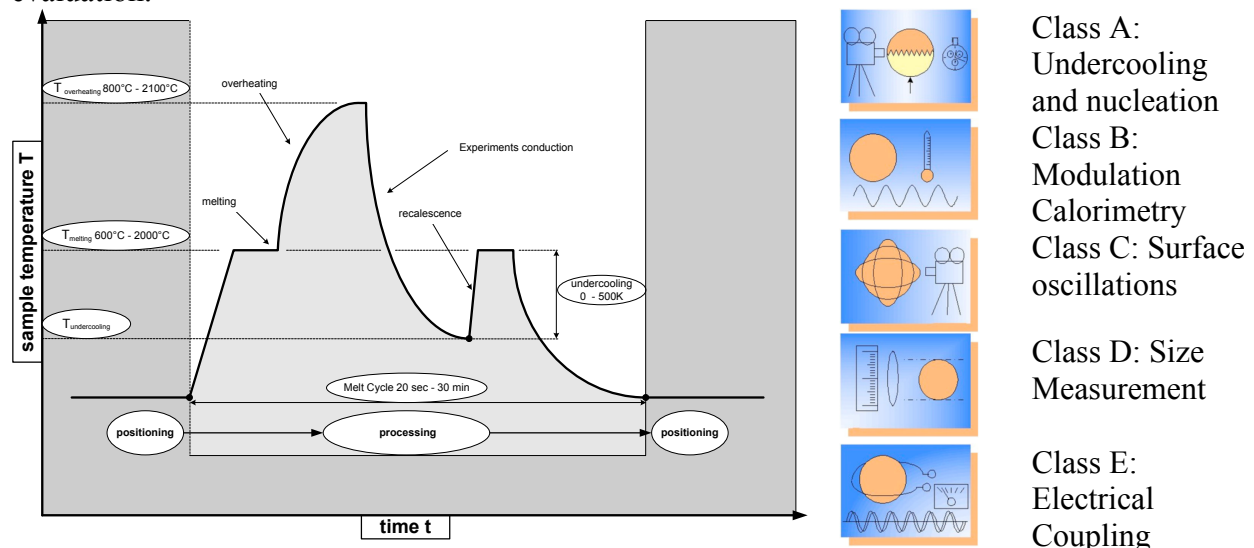


Figure 2. Principle sketch of the temperature-time profile of a typical melt cycle in the EML facility. Experiments are typically performed on the cooling flank when the sample is in the liquid state. Experiments in 5 different classes can be performed with EML, these classes are schematically introduced on the right side of the figure, and explained in detail in the text below.

4. EML on ISS: Experiment Classes and Measurement Capabilities

Based on a long and successful heritage of electromagnetic levitation facilities on various carriers and missions, the electromagnetic levitation facility EML for the International Space Station ISS is currently being developed by Astrium under contracts to ESA and DLR. A comprehensive discussion of the performance and design features of EML is provided in [8].

4.1. Experiment Class A: Undercooling and nucleation

The scientific objectives for this experiment are to:

1. measure solidification front speed
2. perform nucleation statistics (maximum undercooling)
3. study morphology and phase selection depending on undercooling and induced fluid flow [9]

An experiment of class A type is being executed as follows: The molten sample will be cooled by turning off the heater, whereas the cooling rate may be enhanced by dedicated convective cooling per adjustable gas flow. Recalescence may occur either spontaneously or it can be triggered by touching the undercooled sample with a custom-made trigger needle at a predefined undercooling temperature. The recalescence of the undercooled sample is detected automatically by an algorithm that detects the sudden increase of the pyrometer signal upon recalescence. A high-speed video camera is used to observe the sample from a direction perpendicular to the trigger needle such that the nucleation front passing from the nucleation point (trigger needle tip) across the surface of the sample can be observed. Once recalescence has been detected the high-speed video acquisition is stopped. Pre- and post-event storage of video data is facilitated by the use of a ring memory for the video data. See also the first pictogram in figure 2.

Typical duration of one melt cycle is in the range of 30-60 seconds. The temperature is recorded with a frequency of 100 Hz; video images are taken with up to 30.000 frames/s @ 256x256 pixel spatial resolution. An example of high-speed video recording is shown in figure 3.

The following equipment is needed for the execution of a class A experiment:

- high-speed and high resolution camera to resolve solidification process, this includes high digitalization of the camera chip to support good thermal resolution of the phases solidifying
- trigger needle to induce nucleation at desired temperatures of the undercooled liquid
- precision pyrometer

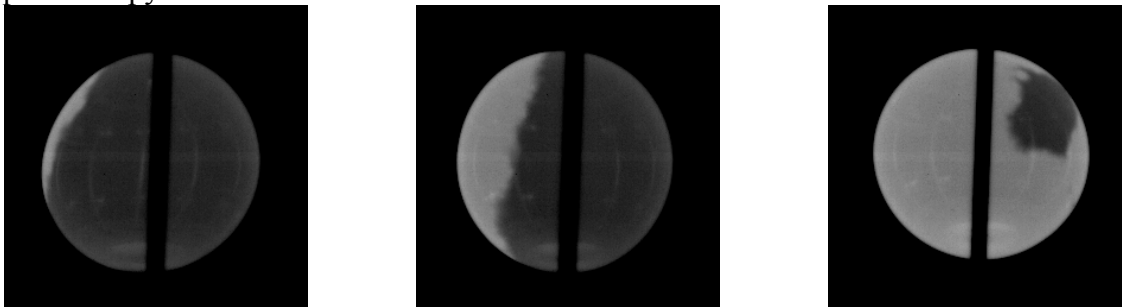


Figure 3. Class A Experiment: Sequence of a NiAl sample solidifying from the undercooled liquid captured by high-speed video camera @ 10.000 frames/s during parabolic flight. The total duration of the recalescence event was 29.1ms.

4.2. Experiment Class B: Modulation Calorimetry

The scientific objectives for this experiment class are to measure thermo-physical properties and if applicable their temperature dependence [10] such as:

1. heat capacity
2. enthalpy of fusion
3. total hemispherical emissivity
4. thermal transport coefficients
5. fraction solid

The class B experiment is carried out as follows: At a constant base temperature of the molten sample the heater power is modulated sinusoidally and the sample's temperature response is evaluated. The modulation can be repeated at different sample temperatures with different modulation frequencies in order to obtain several data points within one melt cycle. The typical duration of a melt cycle ranges from 10 to 20 minutes. The temperature data will be acquired with 100 Hz; furthermore input data of RF coil system (voltage, current) are needed.

The following equipment is mandatory to perform a class B experiment:

- high precision pyrometer
- calibrated modulated power input source (amplitude or power modulation is possible)

4.3. Experiment Class C: Surface Oscillations

The measurement of surface tension and viscosity and their temperature dependence are the main scientific objectives for these experiments.

The experiment starts with heating up the sample slightly above its melting point. During subsequent cooling of the molten sample a short heater pulse is applied which excites surface oscillations due to the dipole nature of the heating field, and the radial force of the field squeezing the sample. The surface oscillations are imaged by two video cameras in orthogonal views, as shown in figure 4. From the oscillating frequency the surface tension can be derived and from the oscillations decay the viscosity can be derived since this is the only damping mechanism involved for these oscillations. The temperature as well as the stimulus (pulse amplitude & shape) can be altered to achieve various measurement points during one melt cycle. For increasing viscosity the amplitude can be increased to observe a high enough oscillation signal. Instead of an excitation pulse also a frequency sweep can be applied to find the resonance frequency of the sample's surface oscillations. The typical duration of a melt cycle is in the range of 5-15 min.

The video images are usually taken with the radial camera @ 200 frames/s and 600x600 pixel spatial resolution and with axial camera @ 150 frames/s and 350x350 pixel spatial resolution. An example is shown in figure 4. The temperature is recorded with 100 Hz.

To perform a class C experiment the following equipment must be provided:

- fast and high resolution video cameras to resolve shape oscillations such that offline Fourier analysis on the sample diameter can be performed to derive the oscillation frequency and decay time
- high precision pyrometer
- free programmable power input source to apply either short pulses or a user defined signal shape as e.g. frequency sweep

4.4. Experiment Class D: Size Measurement

The scientific target for this class of experiments is to measure the thermal expansion coefficient and its temperature dependence. It is carried out as follows: The diameter of the solid and molten sample is precisely determined by the two video cameras in orthogonal view along the changing temperature of a melt cycle. For this purpose a high spatial resolution of the camera is necessary which will be even enhanced by a sub-pixel resolution algorithm which is applied to the video images off-line during evaluation. The algorithm requires the image of the sample's edge to be smeared out over a small number of camera pixels such that the algorithm can fit a 50% intensity value, as assumed sample edge, with a precision higher than the pixel resolution. The slightly blurred imaging of the sample's equator is achieved by setting the cameras focal plane slightly in front of it still achieving a sharp picture of the sample's surface. The typical duration of a melt cycle is in the range of 1-5 min. Video images captured by the axial camera are taken with typically 25 frames/s at up to 1000x1000 pixel resolution, whereas the radial camera offers

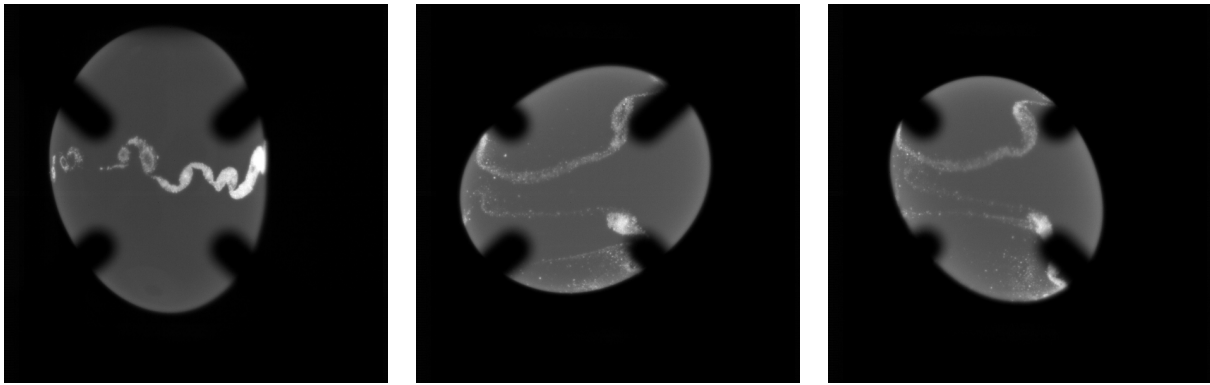


Figure 4. Class C experiment: Induced surface oscillations of a CuSnP sample (TEMPUS, 600x600x pixels @ 200 fps)

600x600 pixel spatial resolution. The selected frame rate depends on the temperature gradient and the desired temperature resolution of individual measurement points. The temperature data are recorded with 100 Hz.

The following equipment is needed for the execution of a class D experiment:

- high resolution video cameras to resolve sample diameter
- high precision pyrometer

4.5. Experiment Class E: Electrical coupling

The measurement of the electrical conductivity and its temperature dependence are the scientific targets for these investigations.

A class E experiment is carried out as follows: The electrical data of the RF oscillating circuit are used to derive the electrical conductivity of the sample since the sample itself is changing slightly the characteristics of the RF oscillating circuit by its inductance, which is determined by its geometry and electrical conductivity. Some calibration measurements on the empty coil have to be performed to distinguish between the coil's inductance and the one introduced by the sample. The relevant housekeeping data are measured by the levitation power supply which uses them mainly for internal control of the high RF power output. The basic resolution and accuracy can be further enhanced by using dedicated measurement equipment for this task- the so-called SCE which is described later.

The measurement can be performed together with other experiment classes since it requires only the high precision pyrometer as diagnostics, as all the other experiments too, and the RF oscillating circuit data which are part of the regular EML housekeeping data anyway. The acquisition frequency for the temperature data is 100 Hz, the RF oscillating circuit data (voltage, current, frequency) are recorded with 10 Hz.

5. Experiment preparation

Experiment preparation involves all aspects of transforming a scientific objective or idea into a fully developed flight experiment. Since EML provides a wide range of functions related to the definition of the sample processing including diagnostics, a deep understanding of the associated systems, their capabilities, functions, interrelations and constraints is needed in order to develop a successful flight experiment. Support to the scientists for the preparation of their experiments is given in the frame of the Ground Support Program GSP by a joint team consisting of Astrium and the DLR Microgravity User Support Center MUSC. Thereby the experience made in former TEMPUS missions is fully provided to the scientific community. A brief outline of the support tasks is given hereafter. The GSP is described in more detail in [12].

From the EML facility operation point of view the experiments are organized into melt cycles which in turn are decomposed into process steps. Each process step is characterized by a number of EML facility settings such as heater power, camera speed, etc. These settings are called Experiment Parameters or EP. In order to perform an experiment in the EML facility all EPs belonging to this experiment are needed since the EML facility is being operated using these parameters.

One of the major tasks of the GSP is therefore to translate an experiment idea into an EP set, a task that involves multiple steps and activities (see also figure 5), such as

- Definition of a rough outline of the experiment, in a so-called "science protocol"
- Characterization of sample properties relevant for processing in EML in a ground levitator
- Development of the Experiment Parameter (EP) set
- Verification of the EP set in a representative EML facility on ground
- Sample Integration and Launch

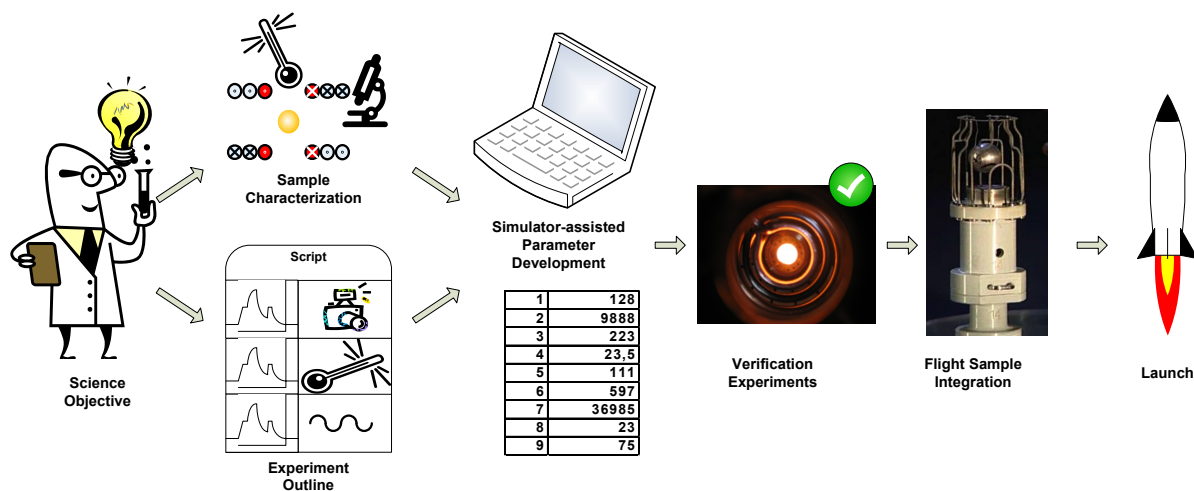


Figure 5. Preparation tasks for conducting experiments in EML onboard the ISS.

5. Mission Scenario

The EML facility will be launched and transported to the International Space Station ISS, where it will be installed in the European Drawer Rack (EDR). During its service lifetime of 5 years EML will be operated in the EDR. In order to be able to process a large number of scientific samples during the service lifetime, samples can be uploaded and downloaded between ground and EML on ISS according to the following scenario (see also figure 6):

- The flight samples which include pure metals, alloys and semiconductors, are spherical in shape with diameters ranging from 5 to 8 millimeters and are manufactured by the participating scientists in their labs.
- These samples are then integrated into dedicated sample holders and subsequently into a Sample Chamber (SCH) under very strictly controlled environmental conditions since the scientific results of the levitation experiments depend critically on the purity of the samples. Each Sample Chamber has a capacity of 18 samples which are stored in individual sample holders mounted on a magazine wheel in order to manipulate and select each sample individually for processing.
- The fully loaded Sample Chamber is uploaded to the ISS where it is attached to the EML facility.
- Sample processing is performed according to a sophisticated procedure which not only involves the settings of the levitator but also of diagnostics elements. The data delivered by the diagnostic

elements, mainly high-speed and high-resolution video cameras and pyrometers are the main scientific raw data gathered during the mission. Since the duration of a typical melt cycle of an EML sample is in the order of seconds or minutes and the processing procedure is very complex with hundreds of individual settings, the procedure is developed before the mission in a ground based experiment development program, converted into a parameter set for the facility, and is then automatically executed by the facility. Interactive control of running experiments can be performed e.g. to optimize parameters based on the results of the previous runs by tele-commanding from the user center on ground which is permanently monitoring and controlling the running experiments.

- After all samples in a Sample Chamber have been processed they are transferred to a Sample Download Container (SDC) by the flight crew for download to ground. The design of SDC has been optimized for the tight constraints of the download in terms of mass and size.
- After landing the processed flight samples are removed from the SDC and returned to the scientists for further analysis.

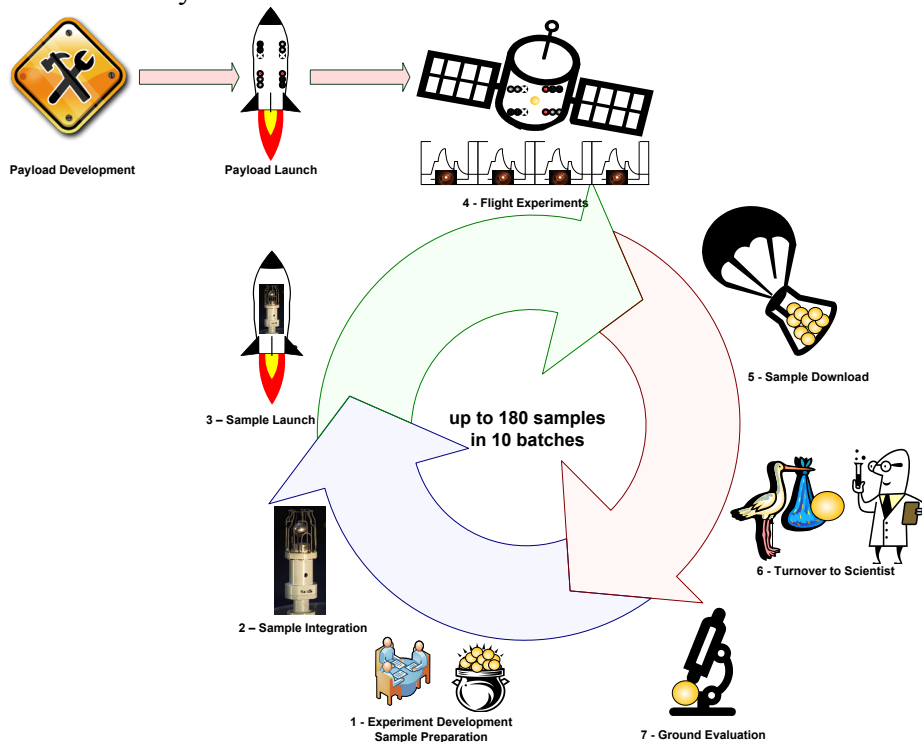


Figure 6. EML Mission Scenario, showing the complete life-cycle of the experiments from experiment preparation to post-flight evaluation.

According to the mission scenario up to two Sample Chambers per year will be uploaded to the EML facility, which translates to a total of 36 samples that can be uploaded, processed, and downloaded per year. Therefore, a maximum of 180 samples can be processed during the 5 year service lifetime of EML.

6. Advanced experimental techniques

The scientific outcome of the experiments as described herein may be significantly improved by implementing the following two upgrades of the EML facility [1], [9], [13], [14]: Oxygen Control System (OCS) & Sample Coupling Electronics (SCE). A description of these instruments and their capabilities is provided in [15].

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