MARS, a new beamline for radioactive matter studies at SOLEIL

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Abstract. MARS (Multi Analyses on Radioactive Samples) beamline is the hard X-ray bending magnet beamline dedicated to the study of radioactive matter of the new French synchrotron SOLEIL. The beamline, which has been built thanks to a close partnership and support by the CEA, has been designed to provide X-rays in the energy range of 3.5 keV to 35 keV. This allows to encompass M and L absorption edges of actinides, as well as K edges of transition metals (that are present in alloys and fuel claddings) up to heavy halogens, rare gases and alkalis (fission products in nuclear fuels). The MARS project aims to extend the possibilities of synchrotron based X-ray characterizations towards a wider variety of radioactive elements and a wider variety of techniques than what is currently available at other facilities. Thus, its specific and innovative infrastructure has been optimized in order to carry out analyses on materials with activities up to 18.5 GBq per sample for α and β emitters and 2 GBq for γ and n emitters. So, today, more than 70 different elements and more than 350 different isotopes have been proposed for studies on the beamline by the involved user community. The arrangement of the different elements in the optics hutch is based on an original scheme which permits to have two alternative optical configurations (monochromatic or dispersive) depending on the nature of experiments to be performed. At least three main techniques are progressively being proposed on the three complementary end-stations located in the experimental hutch: transmission and high resolution powder diffraction (TXRD and HRXRD), standard and dispersive X-ray absorption spectroscopy (XAS and EDXAS) and X-ray fluorescence (XRF). In addition, by using the KB optics, a micro-focused beam will be available on the second station of the monochromatic branch. The beamline is currently under commissioning. The first two experimental stations, using the monochromatic branch, are scheduled to be operational at the end of the year.

1. Source and infrastructure
MARS beamline is built on the bending magnet port D03-1 of the SOLEIL storage ring. The 1.71 T bending magnet field provides a continuous spectrum of photons with a critical energy of 8.6 keV. Due to the particularity of the samples to be studied and the high radio-activities foreseen, the beamline is characterized by a specific innovative infrastructure separated into three main functional areas (see figure 1). The first and the last area, which are standard, correspond respectively to the Pb shielded optics hutch and to the main control room. The central area, which is a restricted zone under authorization because of the presence of radioactive samples, includes essentially the experimental hutch, the sample conditioning and storage room and a secondary control room. It is in this area that special infrastructural solutions have been realized in order to guarantee safe activities with radionuclide samples. Indeed, in order to guarantee a dynamical confinement in case of a
contamination incident, all the area is set under depressurised atmosphere (-80 Pa). Thus, a dedicated ventilation system, with special absolute filters, as well as special air-tight wall internal layers and cables feed-throughs, have been installed. Also, all the internal infrastructural elements of this area have been constructed using materials which are fireproof, with a degree of resistance of 2 hours, and which can be easily cleaned in case of contamination [1].

**Figure 1.** General layout of the MARS beamline from the optics hutch (right side) to the control room (left side). The central part is the restricted area for radioactive sample preparation and experiments.

### 2. Beamline optics

The main characteristic of MARS optics is its original scheme which permits to set two alternative optical configurations (monochromatic or dispersive) depending on the nature of experiments to be performed (see figure 2). The main optical elements, which accept a maximum horizontal fan of 3 mrad coming from the bending magnet radiation, are two long mirrors (M1 and M2) and two independent monochromators, a double crystal monochromator (DCM) first and a single bent crystal monochromator (SCM) downstream. To switch from the monochromatic mode to the dispersive mode, the DCM is moved out of the beam and the height of the M2 is reduced to a value depending on the glancing angle of both mirrors [2].

The optical elements have been designed taking into account the characteristics of the bending magnet source, in order to reach, in the monochromatic configuration, the maximum of the beam flux inside the energy range of 3.5 to 35 KeV (with $10^{12}$ ph/s at 12 keV), and with the possibility of focusing the beam to a minimum theoretical size of around $100 \times 100 \mu m^2$. Furthermore, micro beams with dimensions below $10 \times 10 \mu m^2$ will be available on one of the experimental stations, by using of a set of mirrors in the Kirkpatrick-Baez (KB) geometry. Beyond the scientific interest, these focused modes on both horizontal and vertical planes provide a means to decrease the volume of analyzed materials and consequently the effective dose rate from samples.

The mirrors, manufactured by IRELEC and SESO, provide respectively, a vertically collimated and focused beam. To optimize the flux on the sample over the entire energy range, the mirrors consists of silicon blocks with two different reflective surfaces: a 60 mm width silicon uncoated strip for energy setups below 14 keV and a 50 mm width platinum coated strip (with a thickness of 60 nm) for high energy. Because of the limited length of the mirrors, the minimum glancing angle was fixed to 2 mrad in order to get reasonable photon fluxes at high energies. Both mirrors are operated in ultra high vacuum and are water cooled by copper blades inserted into grooves (via gallium eutectic) because of the heat load coming, respectively, from the direct white beam for M1 and from the band-pass filtered beam in the dispersive configuration for M2. Prior their installation, optical tests showed that the slope
errors for M1 and M2 are equal to 1.5 µrad and 1.0 µrad RMS respectively and the roughness of both mirrors is around 0.25 nm (values obtained along the 1200 x 110 mm$^2$ active surface).

**Standard configuration**

![Standard configuration diagram](image)

**Dispersive configuration**

![Dispersive configuration diagram](image)

Figure 2. Schematic representation of both configurations of MARS optics showing the main components and their respective distances from the source.

The double crystal monochromator (DCM), manufactured by FMB Oxford, is equipped with 14 independent motorized stages in order to optimize its alignment and then to assure a fixed exit beam over the entire energy range with a constant upward offset of 20 mm with respect to the primary beam. Optical alignment of the crystal Bragg axes was done with a particular attention because of its criticalness. The DCM is also equipped with a bender device on which the second crystals are mounted to perform the horizontal dynamic focusing of the beam on the sample. The original feature of this DCM, designed for relatively large beams (H x V = 30 mm x 5 mm), is its ability to exchange crystal sets under vacuum, i.e. from Si(111) crystals to Si(220) crystals and vice versa. By this way, such operation including realignment can be done in less than 30 minutes without venting the vessel. Both second crystals can be also bended sagittally in order to focus the beam in the horizontal plane.

The single crystal monochromator (SCM), designed by SOLEIL, is integrated in a common vacuum vessel with the beam shutter and is installed after the second mirror. The SCM is switched on by a single vertical translation for the dispersive station. The energy dispersion and the horizontal focusing will be achieved by Bragg reflection on the elliptically bended Si(111) or Si(311). The dispersive configuration will be mainly selected for time resolved experiments (ms regime) or low resolution (on consequently high flux) characterizations in the energy range of 5 – 23 keV. A focused beam size of 11 x 60 µm$^2$ with 10$^{12}$ ph/s should be obtained at 12 keV. The SCM as well as the dispersive XAS station will be installed in a subsequent construction phase of the beamline (currently foreseen in 2010 or 2011).

3. Experimental stations and commissioning

Excluding the dispersive XAS station, two end stations located on the monochromatic branch of the beamline are being progressively installed and commissioned in the experimental hutch (see figure 3).

The first is a high-resolution diffraction station which mainly consists of a robust two-circle diffractometer which is installed 23 m downstream the X-ray source. An inner sample-holder goniometer (Khi and Phi circles and x,y,z stages) and a multicrystal analyzer detection system have been also designed and will be added to the diffractometer before the end of 2009. Improvements in the design and characteristics of these latter two equipments have been made thanks to a large implication and participation of CEA.
The second station is dedicated to standard X-ray absorption spectroscopy measurements as well as X-ray fluorescence and transmission X-ray diffraction measurements. It consists on a highly rigid experimental table with a honeycomb breadboard support and a motorized base with micrometric vertical elevation. Samples are positioned in the central part of the board at 26 m from the X-ray source. At present two different sample holders have been manufactured: a special multi-axis motorized stage and a He flow cryostat to allow measurement down to T=10 K. The different intensity monitors, experimental slits and reference sample-holder are supported on 2 aluminum rails which are positioned on either side of the sample. Apart from intensity monitors for transmission XAS detection (in-house device based on photodiodes), several different detection systems are being progressively installed on this station: an imaging plate scanner detector (from Marresearch GmbH) has been purchased; a multi-element Ge detector as well as a SDD detector are foreseen for X-ray fluorescence measurements; an in-house X-ray camera has been manufactured by SOLEIL’s Detectors Group. Finally, as mentioned before, it is also foreseen to mount on this experimental station, a KB mirrors optics system. This will extend the performances of the beamline towards micrometric scale characterizations such as microXRF mapping, microXAS and possibly microXRD.

![Figure 3. Layout of the experimental stations using the monochromatic branch of the beamline (with distances from the source). Left: optical elevating table for standard X-ray spectroscopy; Right: two-circles diffractometer for high resolution X-ray diffraction and beam conditioning stage.](image)

Since September 2008, the MARS beamline commissioning is progressing, starting from the optics up to the end stations. Currently, first test experiments with expert users have been conducted on the standard absorption station with non radioactive samples. First XAS as well as transmission XRD measurements have been performed (also by using different sample environments like furnace or high pressure cell). Opening to external users is scheduled for first semester 2010 only for experiments on samples with a radioactivity below the French exemption limit. Experiments with radioactive samples above the exemption limit will take place as soon as the French government authorization to receive radioactive samples will be given (currently, licensing is expected by the end of 2010).

References

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[2] Sitaud B and Lequien S 2006 Proc. Work. on Speciation Techniques and Facilities for Radioactive Materials at Synchrotron Light Sources (Berkley) (NEA vol. 6046) pp 71-79