NEW SPECIMEN HOLDER FOR XAS-ANALYSES OF RADIOACTIVE SPECIMENS AT THESWISS LIGHT SOURCE (SLS)

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ABSTRACT
A new specimen holder for the safe containment of radioactive specimens has been developed in PSI. This holder will allow XRay Absorption Spectroscopy (XAS) analyses of metallic or liquid low level activity materials at the Swiss Light Source SLS. The holder insures a tight containment of the specimen with two to three layers of Kapton foils for the X-Ray transmission. The holder design and the loading procedure that will be used to insure a contamination free external surface will be described and discussed. Preliminary measurements realized during the commissioning of the beam line with the new specimen holder will also be presented.

KEYWORDS **Holder, XAS**

1. INTRODUCTION

The Paul Scherrer Institut (PSI) is commissioning a new beam line for X-Ray Absorption Spectroscopy (XAS) with a micrometer beam size on the Swiss Light Source (SLS). This beam line has been partially financed and built by the Nuclear and Safety Department (NES) of the PSI.

The main interest of the NES is to realize micro-analyses of radioactive materials in the main research fields of the department. In order to realize these analyses, a special holder insuring the safe containment of the specimens had to be designed and constructed.

This paper contains a presentation of the main features of this specimen holder.

2. THE SWISS LIGHT SOURCE AND THE µ-XAS BEAM LINE

The Swiss Light Source (SLS) at the Paul Scherrer Institut is a third-generation synchrotron light source. With an energy of 2.4 GeV, it provides photon beams of high brightness for research in materials science, biology and chemistry. At the moment 6 beam lines are fully operational and 6 other at different stage of planning, construction or commissioning[1] [1].

The µ-XAS beam line is a new line installed on a Undulator in order to obtain a very high brightness. It will focus on X-Ray Absorption Spectroscopy (XAS) and X-Ray Fluorescence (XRF) experiments requiring high spatial resolution. Furthermore, the beam line will host the FEMTO project that will allow investigations of time-dependent phenomena in the femtosecond time regime. The design of the µ-XAS beam line is conceptualized to yield monochromatic X-ray beams (5-20 keV) with high energy resolution combined with dynamic micro-focusing capabilities[2] [2]. The main characteristics of the µ-XAS beam line are given in Table 1.

Table 1. Main characteristics of the µ-XAS beam line of the SLS

Energy range	5 - 20 keV
Flux on sample	2 x 1012 ph/s/400 mA
Spot size on sample (expected)	1 x 1 µm2
Polarization	linear horizontal / circular left & right
Photon energy resolution	0.02%

The commissioning of the line started early in 2005 and is planned to continue until end of the year with the installation and test of all optical and analytical equipments.
The energy range allows using the K lines or the L lines for the study of the main structural materials (Steel, Zircaloy) or respectively actinide materials (Th, U, Pu) of interest for the NES.

[1] More information can be found in http://sls.web.psi.ch/.
[2] More information can be found in http://sls.web.psi.ch/view.php/beamlines/mxas

The main interest of the department lies in the study of the composition and chemical bounding of different elements in nuclear material using the Extended X-ray Absorption Fine Structure (EXAFS) spectroscopy, and Near-Edge X-ray Absorption Fine Structure (NEXAFS). For example, this will allow the study of the speciation of trace elements in nuclear waste or the detailed local analysis of the corrosion layers of Zircaloy. Such studies are actually not possible on radioactive specimens with the existing instrumentation in Switzerland.

3. PROBLEMATIC – WHY A SPECIAL SPECIMEN HOLDER

The SLS in general and the µ-XAS beam line in particular are not designed specifically for the analysis of radioactive materials. Large areas of the SLS building are accessible to everybody without dosimeter (zones F, C and D on figure 1); therefore the dose rate allowed in these zones is limited to 20 µSv/week. In addition the experimental hutch of the µ-XAS beam line (zone B in figure 1) is designed as a "Zone type 0" meaning that, if higher dose rate is locally accepted, no surface contamination is allowed in this zone. In order to comply with these two constrains, we had to design, develop and construct a new specimen holder and shielding for the measurement of radioactive specimens.

The main difficulty was to design a container with X-ray transparent windows insuring a safe, contamination free measurement of the radioactive specimens. The containment has to be accepted by the nuclear safety authorities for the transfer and measurement of radioactive material, including small fuel specimens, in SLS.
The expected activities of typical specimens that will be analysed on the beam line are given in table 2.

Table 2. Maximal estimated activity of typical specimens to be analyzed on the µ-XAS beam line

Structural material	~100 MBq (60Co, 54Mn)
Zircaloy	~50 MBq (137Cs, 105Ru , 90Sr , 125Sb)
Fuel	~500 MBq (137Cs), ~2 KBq U, ~200 MBq Pu(á)
Waste	1.3 MBq 237Np, 37 MBq 243Am,

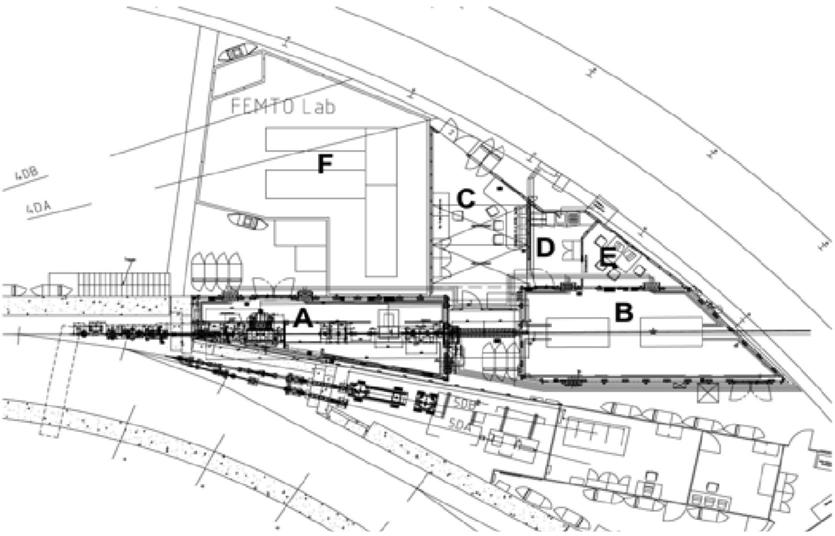

Figure 1. Blueprint of the µ-XAS beam line and surrounding in SLS building

4. CONCEPT DESIGN AND LOADING PROCEDURE OF THE SPECIMEN HOLDER

The specimen holder and surrounding local shielding has been designed to fulfil the following main requisites:

- The specimen holder consists of at least two independent containers
- The specimen is sealed by at least three x-ray transparent layers in the window areas
- The external surface of the external container is contamination free
- The mounting of the specimen in the holder can be realised with master/slave manipulator
- The installation of the specimen holder on the beam line can be realised safely manually
- It is made of light material with low X-ray fluorescence in the energy range of interest

- A dose rate of 20 µSv/h at about 2 m of the specimen in guarantee
- The concept must allow measurements in transmission or in reflection's mode (fluorescence)

The specimen containment system is composed of a specimen carrier holding and fixing of the specimen, a first container named the specimen holder and finally a second container named casing. The windows are sealed with self gluing Kapton® foils. This material is regularly used in accelerators and light sources and has proved its good behaviour under irradiation.

Two designs have been made, one for the measurement of three specimens in fluorescence mode only and a second for transmission or fluorescence modes. They differ only by the fact that the back face of the transmission specimen holder is drilled out in order to allow X-Ray transmission and sealed with an additional Kapton® foil. This paper presents only the specimen holder for fluorescence mode.

First the specimen is glued or embedded on a specimen carrier like presented in figure 2. The specimen carrier is a piece of aluminium alloy of fixed size with a hole adapted to the specimen geometry. It allows a precise positioning of the specimen in the specimen holder. The specimen embedded in the specimen carrier can also be polished in order to get a perfectly plan surface.
The specimen carrier is then put in one of the three available positions in the fluorescence specimen holder

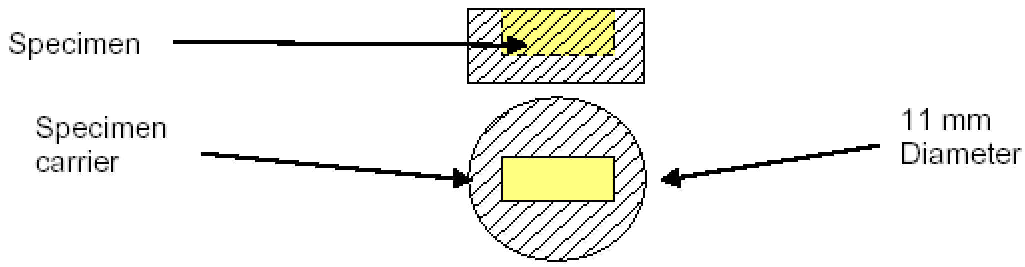

Figure 2. Specimen carrier for fluorescence mode; view from the side (up) and the top (down)

The specimen holder is composed of a base plate with three holes for the specimen carriers and a cover that can be clipped on the base as presented in figure 3. It is built in a radiation resistant plastic named PEEK. The Kapton foil is glued on the cover sealing the three windows in front of the specimens. A YAG crystal is also embedded in the holder and will be used for the fine focusing of the beam. A cap can be mounted on the holder cover for easy manipulation with manipulator and protection during the mounting procedure. The cap will also be used to check the contamination state of the specimen holder after mounting. If the cap is contamination free the specimen can also be considered as contamination free.

At this point the specimen holder can be put on the casing base (figure 4). The casing base is a relative massive piece of aluminium. The specimen holder is positioned on the upper surface at an angle of 45°. The precise orientation of the specimen holder is insured by a pin with corresponding pinhole in the holder. The safety cap of the specimen holder, which is fixed on the cover with a bayonet coupling system, can then be easily removed.

The casing cover is also built in aluminium with the three corresponding windows sealed with Kapton foils (fig. 4). The cover is constructed such a way that the windows can be sealed with two layers of Kapton insuring three layers containment (counting the specimen holder Kapton foil) of the specimen. Again the casing cover is protected by a cap during the mounting procedure. The sealing of the cover is insured by an O-ring positioned between the cover and the base of the casing. The cover is then screwed in position with three screws. At this time the protecting cap can also be removed and checked for possible contamination.

Finally a massive steel cover with a handling bar can be screwed on the holder for the safe manipulation of the holder. This allows an easy transfer of the complete system in the transport cask and from the transport cask on the (X,Y,Z) table of the µ-XAS beam line (fig. 4 right).

The loading of the specimen can be done manually in a hood with light active material or in a shielded box with master/slave manipulator for very active specimens and fuel in particular. In the PSI hot-laboratory, the so called EPMA box chain will be used (fig. 5). The chain is formed by 3 connected shielded cells and one shielded room. The preparation of the specimen and mounting in the specimen carrier will be realised in the first cell (cell 1 in fig.

5). Then the specimen carrier will be transferred in the second cell (cell 2 in fig. 5) where it will be carefully decontaminated with a standard ultra sonic cleaner. After cleaning, the specimen carrier is transferred in the third cell (cell 3 in fig. 5) which is basically clean. In this cell the specimen carrier will be mounted in the specimen holder and the specimen holder will be closed. Finally the specimen holder will be transferred and mounted in the casing in the shielded room. At that point different contamination controls will be realised. If the external surfaces of the casing are contamination free, the transport safety cover will be mounted and the fully encapsulated specimen will be transferred in the transport cask for transfer to SLS.

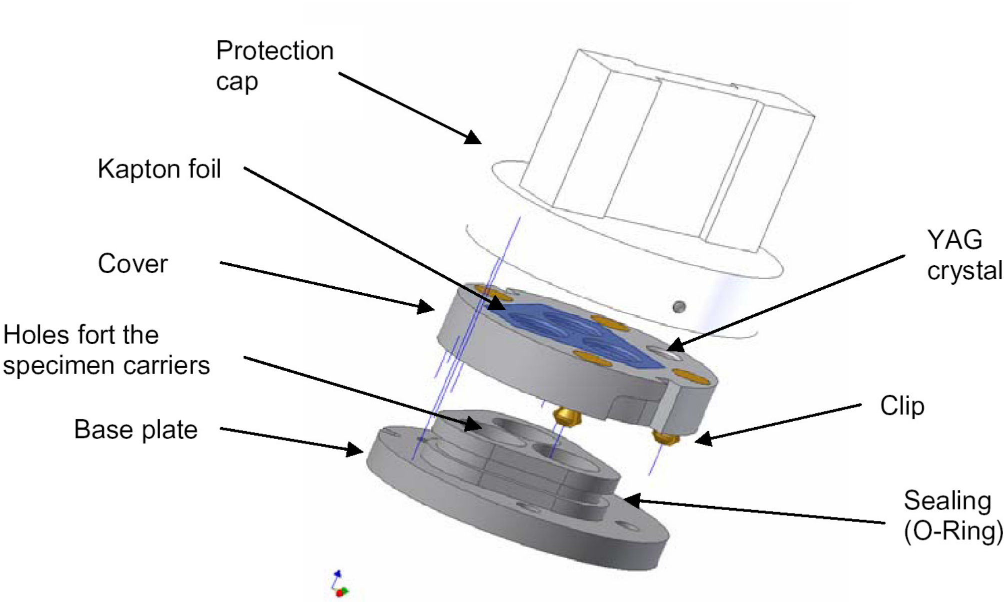

Figure 3. Specimen holder (fluorescence mode) with its protection cap

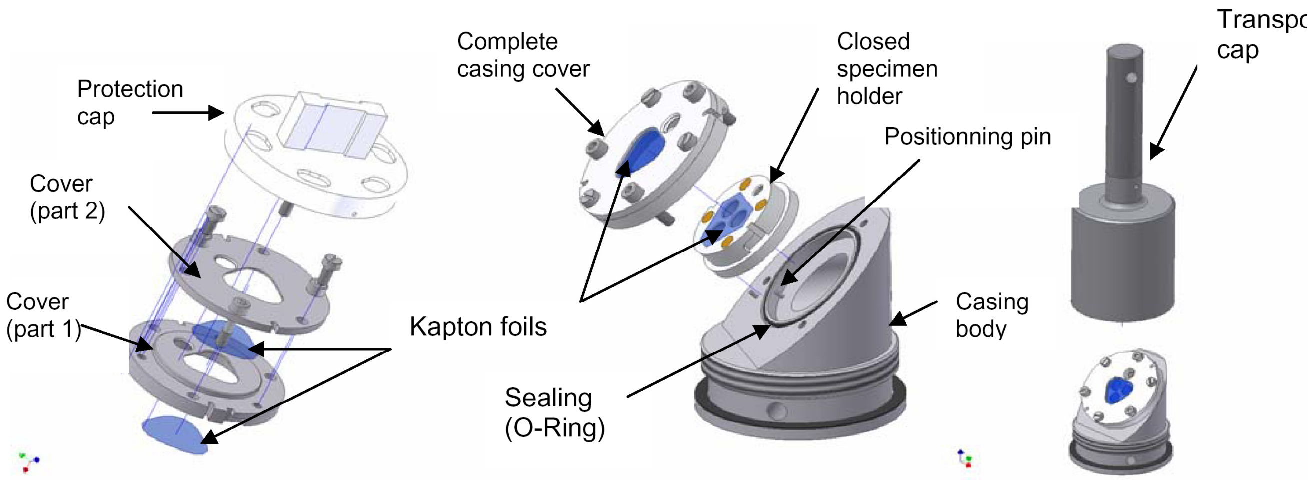

Figure 4. Detail of the casing cover (left) and sketch of the casing with the closed specimen holder (center) and complete holder and casing with the transport safety cap (right)

Finally, a massive steel cover with a handling bar can be screwed on the holder for the safe manipulation of the holder. This allows an easy transfer of the complete system in the transport cask and from the transport cask on the (x,y,z) table of the μ-XAS beam line.

In the SLS, the full system with the safety transport cap will be installed manually on the (x,y,z) table of the μ-XAS beam line. If needed, a circular shielding can be installed around the specimen as it is sketched in figure 6. When the casing is safely fixed on the table the transport safety cap can be removed and the holder is ready for the measurement (figure 6).

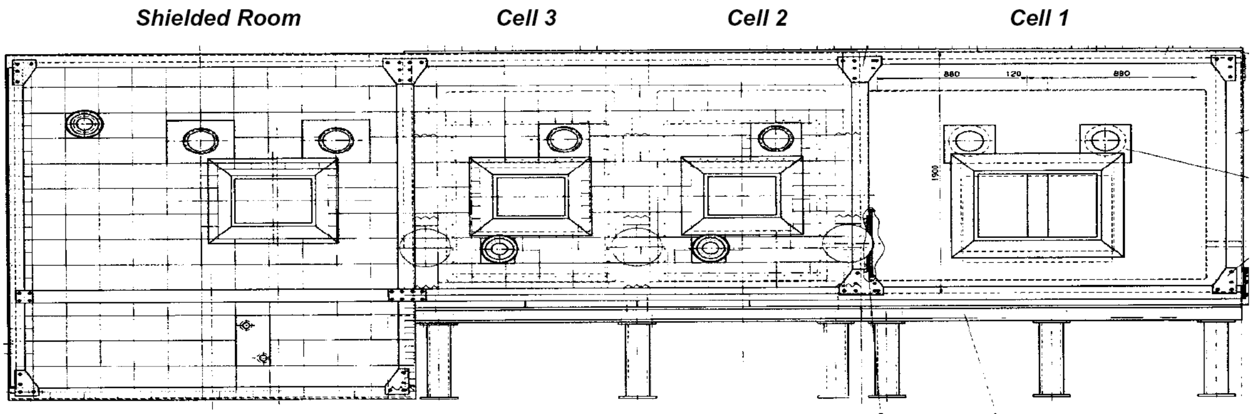

Figure 5. Sketch of the hot cells that will be used for the specimen preparation and mounting in the XAS-containment system.

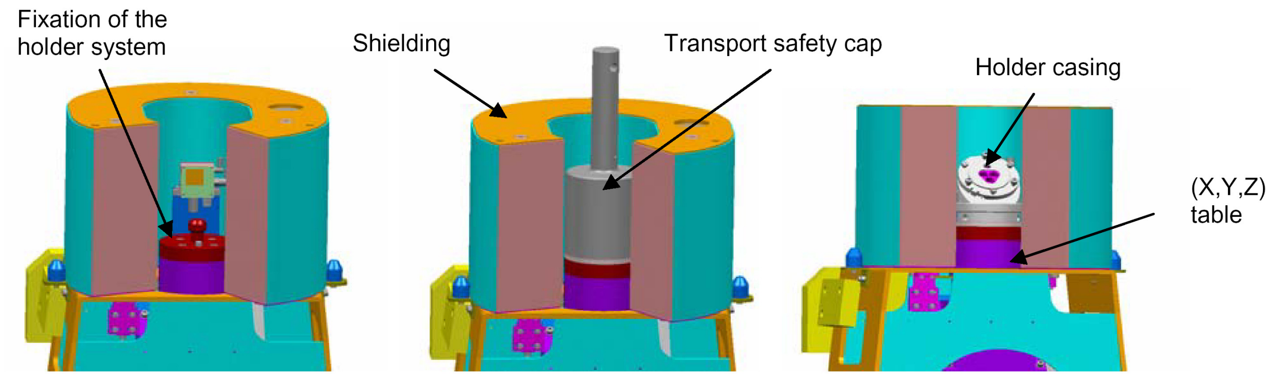

Figure 6 Sketch of the shielding arrangement around the specimen in the µ-XAS hutch without the specimen holder and the transport safety cap (middle) and without the transport safety cap (right).

The complete µ-XAS set up is sketched on figure 7 with the detector support and the extra shielding that will be installed if necessary in order to shield the front direction that is covered by the local shielding around the specimen (beam path).

5. FIRST TEST OF THE SPECIMEN HOLDER

The specimen holder has been built and will be tested with inactive specimens during the commissioning of the beam line
(started in March 2005).

Preliminary tests have been realised with Cu and Zr thin specimens in fluorescence and transmission mode without the (X,Y,Z)-manipulator. The goals of the test were to insure that the containment of the specimen is stable in the X-ray beam, to check that specimen holder geometry allows good measurement and to check the beam transmission through the Kapton foils.

A test of about 30 hours with a millimeter beam has been realised in April 2005. The safe containment of the specimen in the high flux X-Ray beam has been insured through all the test. A microscopic check of the Kapton foils after the test has demonstrated their tightness but the glue layer in the inner surface of the Kapton layers has slightly altered by the X-Ray beam (heating ?). This layer could be removed if needed in the window area.
The test has also proven that the specimen holder and the casing does not modify the characteristics of the incoming beam as it can be seen in figure 8 (left), where the beam intensity distribution for a given energy range is measured with and without the holder (without specimen inside). The spectrum is absolutely not modified by the holder but a small lost of intensity due to absorption in the Kapton layers is observed.

Finally, EXAFS spectrums of Cu and Zr have been recorder in thin specimens. An example obtained on a Zircaloy specimen at the Zr-edge is presented in figure 8 (right). The EXAFS structure is clearly visible, proven the good measurement conditions with the holder.

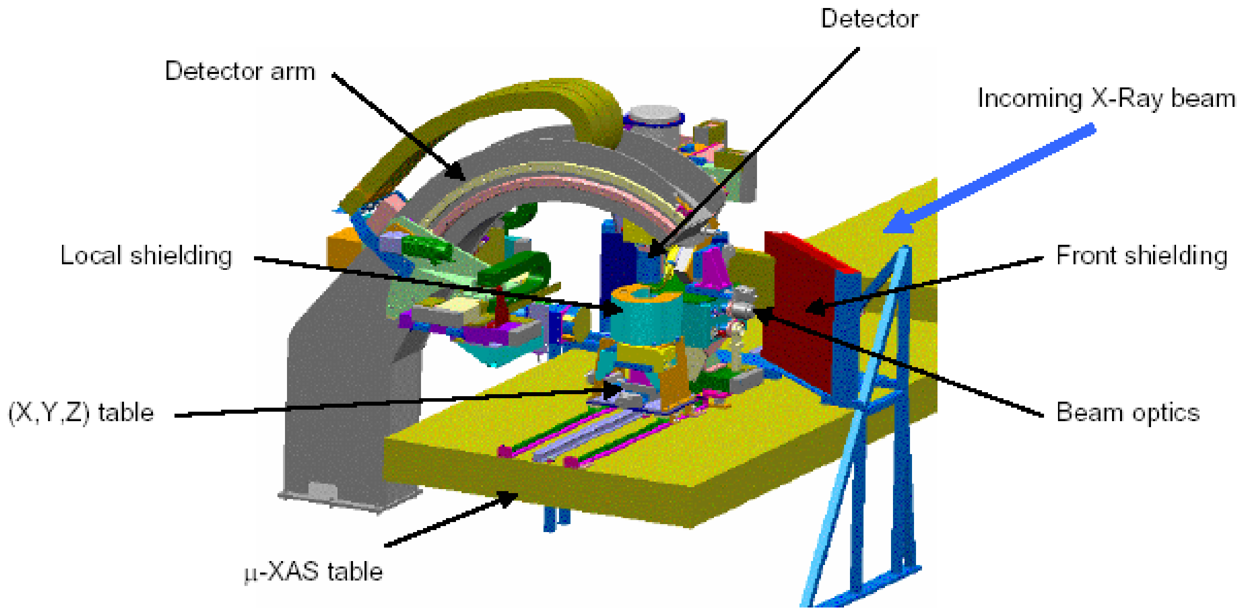

Figure 7 Overview of the complete set up on the μ -XAS table

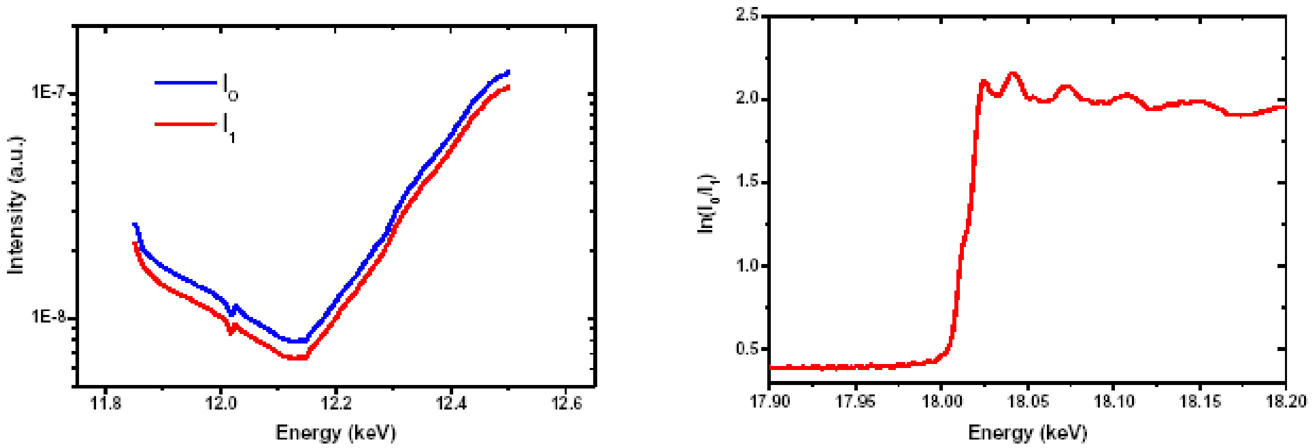

Figure 8 X-ray transmitted intensity of the beam line in the energy range 11.75 to 12.7 keV without (I0) and with (I1) the complete holder system (left) and X-ray transmitted intensity through a zircaloy specimen around the Zr absorption edge - EXAFS (right).

6. SUMMARY

A new specimen holder for the analysis of radioactive material on the XAS beam line at the Swiss Light Source has been designed and fabricated. This holder insures a double or triple containment of the specimen in order to guarantee a contamination free analysis. The loading, transport and transfer procedures have been formally accepted by the safety authorities, but a definitive authorisation will be delivered only after the first test with low activity specimens.

The first tests of the holder demonstrate, that the containment of the specimen is well insured in the beam and that a good spectrum can be recorded in transmission and in fluorescence mode.

The first measurements with light active specimens are foreseen at the end of 2005 after full commissioning of the beam line.