# A versatile beryllium sample holder for amorphous radioactive glasses characterization by synchrotron radiation

*O. Bouty*

Commissariat à l'Energie Atomique, Site de Marcoule, DES, ISEC, DE2D, Univ. Montpellier, BP 17171, 30207 Bagnols-sur-Cèze, France

**Abstract**

We present a beryllium-made device for X-ray diffraction experiments suitable for holding highly radioactive amorphous samples and agreeing synchrotron safety regulations. This container is mainly intended for Wide Angle X-ray Scattering technique, in transmission mode and normal conditions, but can also be extended to other X-ray diffraction methods at different temperature and pressure conditions.

## 1 Introduction

Wide Angle X-ray Scattering (WAXS) is nowadays an easily affordable technique on lab-based diffractometers for characterizing amorphous glasses. It brings out structural information in reciprocal space, and in turn, results in the total radial distribution function in coordinate space when applying an inverse Fourier transform to the data. Accordingly, atomistic environment at short and medium range order can be assessed and compared with the one obtained by atomistic simulation for example. But dealing with highly radioactive amorphous samples introduces many experimental constraints for ordinary usage on lab-based diffractometers, in particular preparation time, specially dedicated controlled areas, glove-boxes, hermetic sample holders, weak signal over noise ratio. Therefore, a balance must be found between sample holder's cost, sample preparation time and characterization. On the other hand, use of synchrotron radiation offers an alternative in terms of high-quality, efficient and quick-time acquisition, in particular with two dimensional detectors. But attending large facilities necessitates respecting many drastic requirements and safety regulations, in particular a double confinement barrier to avoid any potentially radioactive particle leakage.

Several sample holders for radioactive materials characterization in the field of X-ray diffraction already exist in various shapes [6, 2, 5, 1], for which some of them can be used with synchrotron radiation. Precluding unpredictable holders using powder pellets just laid in metallic beds, there are still several drawbacks for the others, one of the most important being the need of a bulk flat polished sample to be stacked in the device, which mainly means a tough preparation, a non-negligible volume and consequently a possibly high or excessive activity with regard to synchrotron acceptable limits. To overcome such constraints, we present a very simple device made with beryllium metal, looking like Russian nesting dolls,

which takes advantage of recent efficient micro-machining techniques for metal-alloys. Our device enables:

- reducing as much as desired the sample's volume and therefore it's radioactive activity (mass possibly less than 10 mg),
- reducing sample's preparation to a very short period of time,
- reducing workforce costs.

## 2 Sample holder

Be is a lightweight element, holds high mechanical and thermal properties and a low X-ray mass absorption factor. It is commonly used in aeronautics and in nuclear field, for example as a power plant neutron moderator. However, Be is hazardous to the health and consequently must be handled with care.

### 2.1 Design

To ensure a perfectly hermetic sample holder, and maximize to the highest safety levels, a set of three Be metal rods with increasing diameters are machined in tubular shape with only one gap. The inner wall of each tube is threaded at the opening to fit with an opposite threaded cap. The inner diameter of the smallest tube is 1.1 mm, with a wall thickness of 0.3 mm. The first, second and third Be tubes are all machined as to fit into one another. A schematic drawing is shown in Fig. 1. The resulting device looks like Russian nesting dolls (Fig. 2).

### 2.2 Leakage test

A leakage test was carried out with the smaller tube filled with tritiated water. The container resulting from three embedded tubes and threaded caps sealed with a spot of Loctite© glue was left for several days in a liquid scintillator. A weak signal edged up from noise only after 20 days, therefore ensuring that the overall device was perfectly safe for a several day lasting experiment in radioactive conditions.

### 2.3 Sample introduction

Sample preparation doesn't need any kind of binder. The glass powder is introduced in the inner tube hold vertically by means of a special funnel-shaped device, held by four legs lying on a base (Fig. 3). A small amount of powder is pushed threw the funnel inside the tube with a plunger. If the operator applies a strength of the order of 10 N by hand on the plunger, then the resulting pressure applied on a surface of 0.9 $mm^2$ (corresponding to a diameter of 1.1 mm) is $10^7$ Pa (or 10 bars) in average. This operation is repeated until the inner tube is completely filled, and finally the cap is screwed and sealed with a spot of Loctite© glue.

### 2.4 WAXS pattern

Depending on the rod fabrication process, Be appears mainly crystallized, with possibly a small fraction of amorphous phase. As the inner Be tube wall is in contact with the sample,

scattered X-rays from the sample will cross a Be thickness in a large angle range, therefore creating an X-ray diffraction pattern overlapping sample's one. This is the main drawback of the device, which must be taken into account during the X-ray diffracted pattern correction procedure. To better assess this contribution, we present an X-ray diffraction pattern of the empty Be holder, achieved at the KAT-ACT beamline located at KARA synchrotron (Germany). The incoming X-ray reached an energy of 50.24 keV and an image plate was used as a detector (Fig. 4). More extensive details of this experiment are described in another paper and readers are encouraged refering to it [3]. If all the beam parameters are unchanged between measurement of the empty Be container and the sample inside the Be container, the following correction procedure holds to obtain a corrected intensity:

$$I_{\mathrm{S}}^{(0)}(\theta) = \left[\left[I_{\mathrm{SCB}}(\theta) - I_{\mathrm{SCB}}^{\mathrm{MS}}(\theta) - I_{\mathrm{SCB}}^{\mathrm{FL}}(\theta)\right]\frac{T_S}{A_S(\theta)} - I_{\mathrm{CB}}(\theta)\right]\frac{T_{\mathrm{C}}}{A_{\mathrm{C}}(\theta)} \tag{1}$$

where $\theta$ is the diffraction angle, $I_{\mathrm{SCB}}$ is the measured intensity from the sample + container + background, $I_{\mathrm{CB}}$ is the diffracted intensity resulting from the empty container + background, $I_{\mathrm{SCB}}^{MS}$ is the multiple scattering intensity and $I_{\mathrm{SCB}}^{\mathrm{FL}}$ the fluorescence generated by the irradiated sample and leaving the container. $A_{\mathrm{S}}$ and $A_{\mathrm{C}}$ are the absorption of the sample and container respectively, $T_{\mathrm{S}}$ and $T_{\mathrm{C}}$ being the transmission factor at normal incidence for the sample and container respectively. Other corrections like beam's polarization or setup geometry ones can be applied afterwards. Determing accurately all these contributions is a key point to a conclusive result.

### 2.5 X-ray absorption

X-ray absorption depends on the container's shape. In cylindrical geometry, analytical formulae are only available at 0° and 90° diffraction angles. Therefore, the best approach to take into account geometry and wall thickness, is to simulate the X-rays propagating through the container towards the detector by means of finite elements and particle-tracing method. An appropriate model is presented by Damay et al. [4]. In this model, air and container are discretised by squared cells in a large array with different ascribed attenuation coefficients. A beam's element enters the array along an axis perpendicular to the detector, moves from one cell to another until scattering occurs in the container, and then follows the new direction until it reaches one of the grid boarders. The transmission of the beam's element is calculated in each cell along its path with the related absorption value. Fig. 5 highlights the very small absorption amplitude variation for an momentum transfer range from 0 to 30 $\mathrm{\AA}^{-1}$, which facilitates the overall pattern corrections.

## 3 Conclusion

A versatile sample holder has been designed to be used with synchrotron radiation and WAXS technique, in transmission mode, at room temperature and atmospheric pressure. The device was succesfully tested with a Cm-doped borosilicate glass known as International Simple Glass (ISG) in the context of nuclear-waste repository research field [3]. Moreover, the container is also compatible with other techniques like Small Angle X-ray Scattering (SAXS) and its use can also be extended to both transmission and reflection modes, and to other temperature and/or pressure conditions.

## References

[1] B. Ao, X. Wang, Y. Wei, and Y. Zhang. A simple hermetic sample holder for X-ray diffraction analysis of uranium hydride. *J. Appl. Cryst.*, 40(4):796–798, 2007.

[2] R. C. Belin, P. J. Valenza, M. A. Reynaud, and P. E. Raison. New hermetic sample holder for radioactive materials fitting to Siemens D5000 and Bruker D8 X-ray diffractometers: Application to the Rietveld analysis of plutonium dioxide. *J. Appl. Cryst.*, 37(6):1034–1037, 2004.

[3] Olivier Bouty, Laure Ramond, Kathy Dardenne, and Jörg Rothe. Two-dimensional Wide Angle X-Ray Scattering on a radioactive glass fitted in a beryllium container. *J. Synchrotron Rad.*, under review, 2020.

[4] Pierre Damay and Abdenacer Idrissi. Evaluation of self-attenuation parameters for neutron and X-ray scattering using a Direct Method. Application to slabs, cylinders and samples of any shape. *Physica B: Condensed Matter*, 382(1):71–75, 2006.

[5] Mark A. Rodriguez, Timothy J. Boyle, Pin Yang, and Damon L. Harris. A beryllium dome specimen holder for XRD analysis of air sensitive materials. *Powder Diffr.*, 23(2):121–124, 2008.

[6] D. M. Strachan, H. T. Schaef, M. J. Schweiger, K. L. Simmons, L. J. Woodcock, and M. K. Krouse. A versatile and inexpensive XRD specimen holder for highly radioactive or hazardous specimens. *Powder Diffr.*, 18(1):23–28, 2003.

Fig. 1: Schematic of the sample holder built upon nested Be tubes. The result looks like Russian nesting dolls

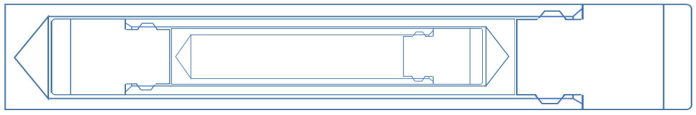


Fig. 2: Be tubes with their threaded caps

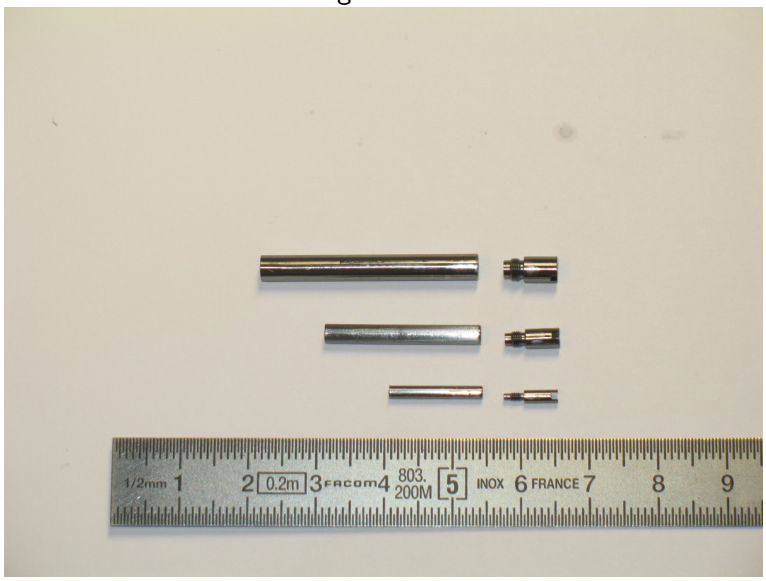


Fig. 3: Funnel-shaped device

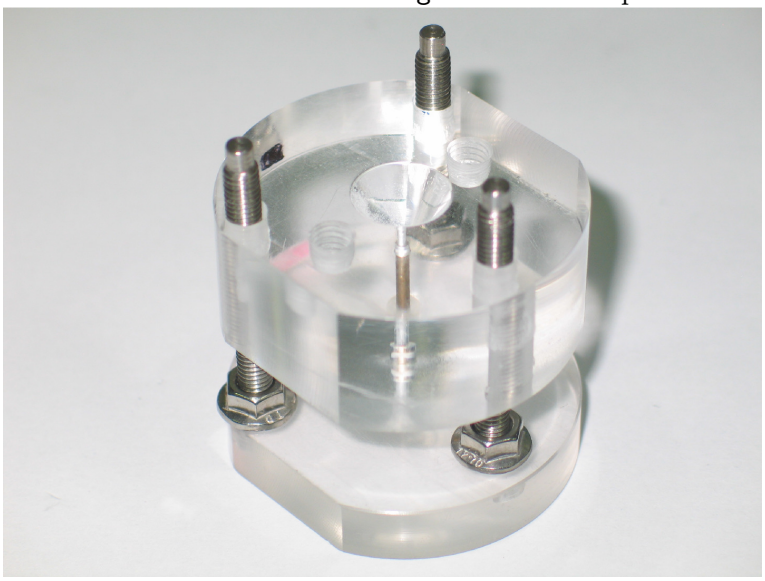


Fig. 4: Be container WAXS pattern

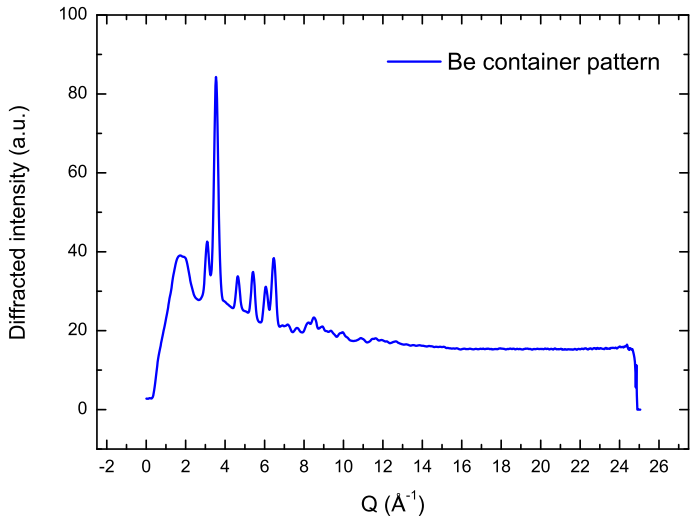


Fig. 5: Absorption from the Be container. Abcissa is refered as momentum transfer (in $\mathring{A}^{-1}$).

