# Development of a specimen containment system and measurement procedures for the neutron diffraction analysis on irradiated fuel materials in PSI

D. Gavillet

Laboratory for Materials Behaviour, Paul Scherrer Institut, CH-5232 Villigen PSI, Switzerland.

## Abstract

PSI is actually operating a large neutron diffraction facility based on a neutron spallation source (SINQ). All spectrometer are installed in a large open hall without the required safety devices for the manipulation of irradiated nuclear fuel.

In order to make a phase analyses of a corium specimen, we have developed a safe container system that can be loaded in the PSI hot laboratory and transported in the neutron diffraction hall without further manipulation of the specimen. A set of safety procedures have also been defined in order to allow the measurement of irradiated fuel material.

The containment system will be fully described and the procedure developed for the contamination control and transport will be explained. Results obtained on a corium specimen will be presented to demonstrate the interest of the method for fuel analysis.

*Keywords*: Containment, Safety, Neutron diffraction, Corium

## 1. Introduction

The Laboratory for Materials Behaviour of the Paul Scherrer Institute got the mandate to study the phase composition of a piece of corium produced in an integral sever accident test (melted $UO_2$ and $ZrO_2$).

The phase composition and geometry of such specimen can be determined with Electron Probe Microanalysis (EPMA). This technique can only provides material analyses near the surface. It is very time consuming to get information on phase distribution in bulk material as it means a large number of polishing steps followed by EPMA analysis. This is why we proposed to realise a neutron diffraction analysis of a small Corium specimen. This technique allows the measurement of large specimens and delivers phase composition and phase distribution and phase structure (crystallography) information on the bulk material.

A high-resolution diffractometer is installed on a measurement line of the spallation neutron source of the PSI (SINQ). The goal of this paper is to present the containment system that has been developed for the transfer of the irradiated fuel specimen from the PSI Hot-Laboratory (HL) to the neutron diffraction hall and the diffraction experiment.

## 2. Description of the problem

The first difficulty is due to the geographic distribution of the PSI facilities. PSI is divided in two sites separated by a river. The hot-laboratory is on the east side and the neutron source on the west side of this river. A bridge connects the two sides but this bridge is open to civil traffic. Therefore, the transport of a fuel piece has to be realised in a certified container.

The second difficulty arises by the fact that the neutron diffraction hall is not prepared and certified for work with specimens containing alpha emitters. It means that no alpha-contamination is allowed in the hall.

The last difficulty is due to the fact that both installations are not under the supervision of the same safety authorities. This has implied to get authorisation from two safety organisations with different regulations.

Therefore, in order to be allowed to realise the measurement on a small piece of corium we had to:

- Develop a safe container that allows the acquisition of a neutron diffraction spectrum and prevent all realistic contamination possibilities.
- Find a certified container for the transfer of the specimen and its containment from the hot-laboratory to the diffractometer.
- Develop a safe procedure for the transfer of the specimen in the containment, the transfer of the containment in the transport shielding, the transfer of the containment on the diffractometer and its return to the hot-laboratory.
- Write a safety report for the specimen transfer and measurement.

This had required more than one year of work and quite some nerves!

## 3. Description of the containment system

The basic idea was to develop a three layers containment system with a strong external container and two internal thin vanadium containers to insure the safe encapsulation of the radioactive specimen. Vanadium was chosen because its neutron diffraction spectrum does not contain large peaks.

We also had to define a loading procedure insuring a contamination free external surface of the containment and this using the existing installation. This in particular limited the possible specimen geometry. In fact we were limited to use stick shaped specimens. The corium specimen is presented in figure 1.

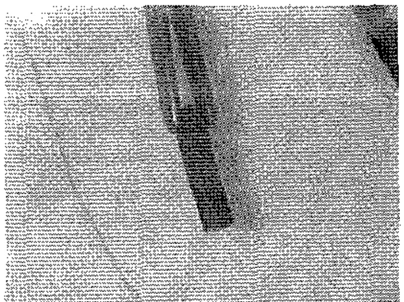

**Fig. 1** Corium specimen (18 x 5 x 3 mm$^3$)

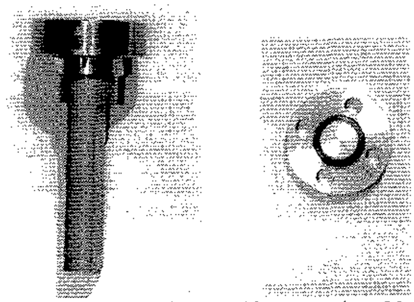

**Fig. 2** First container and its cover (ca. 30 mm height)

The first container has an inner cavity of the size and shape of the specimen and is fully in Vanadium. The cover is fixed with four screws and sealed by an O-ring (figure 2). The specimen is loaded in the containment in a quasi-clean box with a tele-manipulator. When the cover is sealed, the external surfaces of the container are cleaned in an alcohol bath.

The second container has a lower inner cavity of the size and shape of the first container. Its lower part is made in Vanadium. The upper part is in aluminium and was welded on the lower part (figure 3-a). The height of the vanadium part is larger than the vertical size of the neutron beam. The second container is closed with an aluminium cover fixed by a quick vacuum grip system and sealed by an O-ring (figure 3-b). The first container is loaded in the containment in a clean box with a tele-manipulator and its cover is mounted by hand in a closed area on the side of the shielded box. Finally, the external surfaces of the container are cleaned and controlled for contamination.

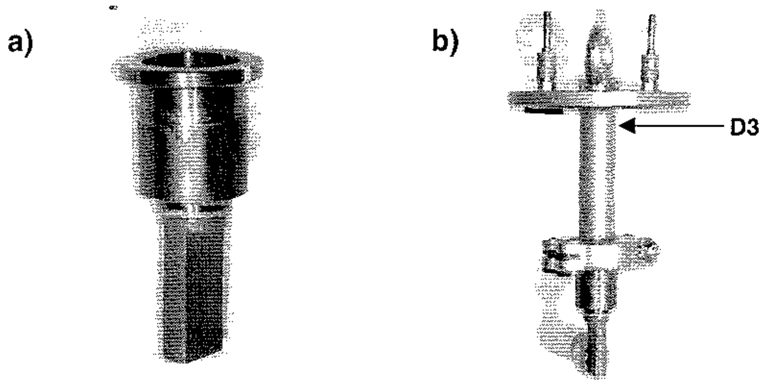


**Fig.3** Bottom part of the second container (a) and the complete container with its cover (b)

The third container is a standard Al tube used for the diffraction measurement. The second container is mounted by hand in the third one and sealed with an O-ring. The head of the second container is used as the cover of the third one (figure 4). The third container is then filled with He in order to decrease the neutron diffraction by air near the specimen.

A test of the empty containment system on the neutron diffractometer has demonstrated that no strong diffraction peaks are produced by this system.

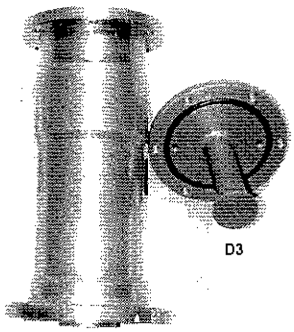


**Fig. 4** Third container with its cover (the upper part of the second one)

At that point the specimen has been ready for transport to the neutron diffraction hall. The safety authorities have accepted this procedure for specimens with a dose rate lower than about 10 mSv/h at 10 cm.

The transport is realised in a large type-A shielded container that is loaded in a small bus.

A complete safety report containing a detailed description of the goal of the experiment, of the containment as well as the loading, transport and measurement procedures and a quite complete evaluation of possible consequences of an accident during the transport and measurement of the specimen including the crash of a plane on the neutron diffraction hall.

After presentation of the safety report and of the tightness test of the container we got the authorisation to realise the measurement on the Corium specimen.

## 4. Measurement and analysis

The specimen is a stick shaped corium piece of 18 x 5 x 3 $mm^3$. Its estimated dose rate was about 10 mSv/h at 10 cm with a U and Pu activity of about 1435 MBq (99% $^{241}Pu/^{241}Am$) and a $\beta\gamma$-emitter activity of about 1000 MBq ($^{137}Cs$, $^{144}Ce$, $^{154}Eu$). After loading, the real dose rate was measured at 6.5 mSv/h at 10 cm confirming the estimation.

The containment system was loaded on the high-resolution diffractometer of the PSI spallation source on a Sunday morning (Figure 5) and the spectrum was acquired for 10 hours.

The neutron wavelength used was 1.8858 Å, the beam was larger than the specimen with a quite flat intensity distribution on the specimen itself. Detail on the spectrometer can be fund on the home page of the SINQ spallation source (www.sinq.ch).

A clear spectrum with a good signal / background ratio was recorded. The small number of peaks indicates a relative homogeneous specimen containing few different phases.

The analysis of the spectrum was realised with standard software developed in PSI. The spectrum is compared with calculated spectra from known crystallographic phases (figure 6).

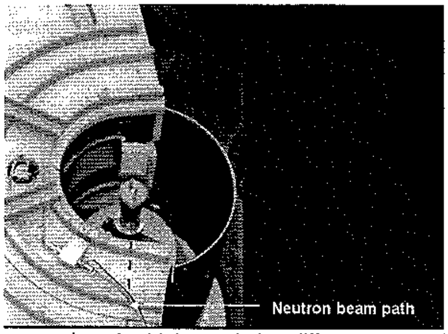


**Fig. 5** Containment system mounted on the high-resolution diffractometer of the SINQ spallation source.

All large diffraction peaks are produced by a single phase with a cubic $UO_2$ type structure and a lattice parameter of about 5.31 Å. This is to compare with the pure $UO_2$ lattice parameter of 5.47 Å and the cubic $ZrO_2$ lattice parameter of about 5.1 Å. This means that the specimen is formed by a $(U_{57\%}, Zr_{43\%})O_2$ compound. This matches well with the other analyses (EPMA and X-Ray diffraction) realised on the same material.

The analysis of the small peaks demonstrates that they are almost all produced by the Vanadium container system.

This proves that the corium is very homogeneous and is composed of a single phase.

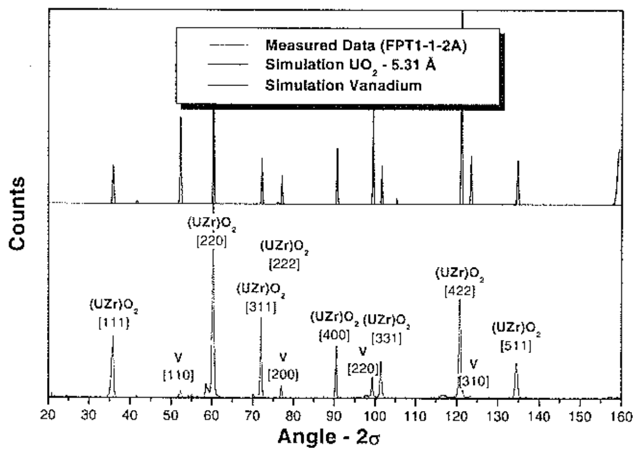


**Fig. 6** Neutron diffraction spectrum acquired on the corium spectra simulation.

## 5. Conclusion

A containment system has been designed, constructed and certified for the transfer and the measurement of fuel specimens on the neutron diffraction spectrometer of the PSI spallation source. The measurement of a corium specimen has demonstrated our capability to realise such tests and also proved the interest of such measurements.

## 6. Acknowledgements

The realisation of this containment system as well as the associated measurement could not have been realised without the participation of a large number of PSI collaborators who are all sincerely thanked. The author would like to thank particularly R. Restani, M. Gehringer and M. Nicolet from the Laboratory of Materials Behaviour, J. Moriawitz, A. Janett, S. Jahn and O. Morath from the radiation control group and L. Keller and P. Fischer from the neutron diffraction group. This would not have been possible without their strong support and help.