

Deadlines for submission of Experimental Reports

- 1st March for experiments carried out up until June of the previous year;



	Experiment title: Structural environment of xenon in silica at high pressures and temperatures (5 GPa-1700 K) by X-ray absorption spectroscopy	Experiment number: HS2272
Beamline: BM29	Date of experiment: from: 7/02/2004 to: 10/02/2004	Date of report: 27/08/2004 <i>Received at ESRF:</i>
Shifts: 9	Local contact(s): S. de Panfilis	
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Report:

The background of this proposal was the missing Xe problem in the atmospheres of the Earth and Mars and the perspective to resolve it by synthesizing Xe-compounds with terrestrial materials. Xe would then cease to behave as incompatible element under high pressures as was shown for Ar (1).

Xe-compounds have been synthesized at ambient pressure from photodissociation of H-containing precursors (2,3). Evidence for the stability of both xenon silicates and xenon hydrates at deep crustal pressures and temperatures was recently presented (4,5) from Raman spectroscopy data.

In situ X-ray diffraction experiments conducted on ID30 on this system considerably improved our understanding of the problem. Over the whole P-T range investigated, 0-5 GPa/300-2000 K, quartz coexists with a liquid. Both phases host xenon. For α -quartz, this is attested by its stabilisation outside of its normal stability field and its volume, systematically larger than expected.

The local environment of xenon meanwhile still remains unresolved although it is a crucial issue of this work. What is the distance to the first neighbors and their number, in the solid phase (α -quartz), as well as in the liquid phase, and most of all, what is the nature of Xe-O interactions and eventually bonds? X-ray absorption spectroscopy is the ideal technique for this purpose especially since we will be able to combine these data with X-ray diffraction data (proposal HS1887) acquired in the same experimental conditions.

The quenched samples contain in general 2-4wt% of Xe in mean but up to 10wt% in some cases.

Mineralogically, they consist mainly of quartz (and coesite for high pressure samples) plus an additional phase, related to SiO₂ clathrates, in which Xe location and atomic environment has also to be found out. In an evaluation of noble gas solubilities in fused silica by molecular dynamic simulations, the silica structure was

also found to be clathrasil-like with heavy rare gases, such as Xe atoms (6). The same conclusion was reached for the solubility of Kr in SiO₂ glass quenched from 0.7 GPa-1570 K and analyzed by EXAFS at the Kr K-edge (7).

Results:

During this run, capsules were leaking, Xe was not retained and therefore absorption data could unfortunately not be recorded *in situ*. Nevertheless, samples quenched from previous experiments were analyzed. Though these samples contain only few wt% of Xe (1 to 4%), the EXAFS signal could be observed (see figure). Since most of the xenon is released from the sample upon T-quenching, this result let us expect much more intense EXAFS signal from *in situ* high pressure/high temperature experiments. In this case indeed, there should be between 20 and 30wt% of Xe in the sample and signal should be consequently amplified.

This run also gave us the opportunity to conceive with BM29 technical staff an efficient way to load Xe in capsules made of relatively transparent material (at the Xe-K edge) and to fix their optimized geometry considering the relative amount of Xe vs. SiO₂. In the particular case of quenched samples, and considering the intensity of the EXAFS signal observed, we might need to add up a fluorescence detector.

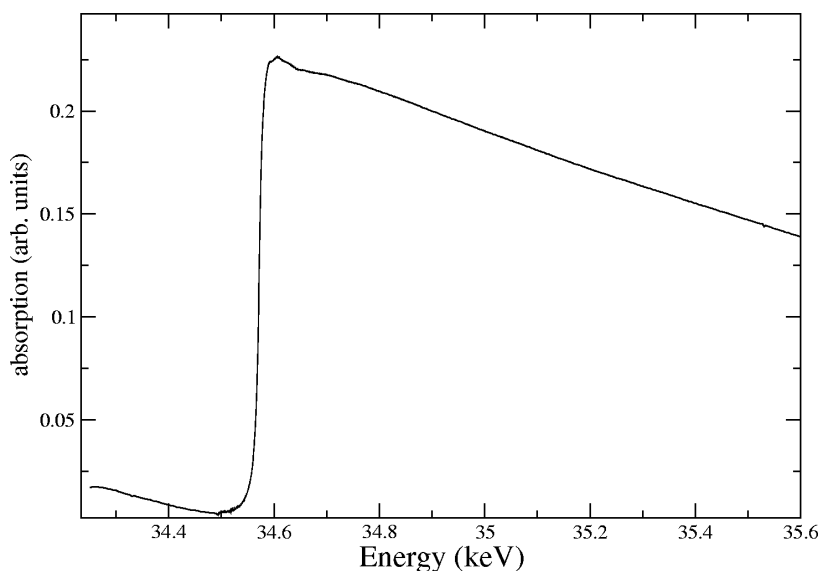


Figure: Absorption spectrum of Xe near the K edge on a quenched sample (run HS2272).

References :

- 1- Chamorro-Perez et al., *Nature*, **393**, p.352, 1998.
- 2- Bartlett, *Proc. Chem. Soc.*, **27**, p. 218, 1962.
- 3- Pettersson et al., *Eur. J. Inorg. Chem.*, **505**, p.729, 1999.
- 4- Sanloup et al., *Proc. Nat. Acad. Sci.*, **99**, p.25, 2002.
- 5- Sanloup et al., *Geophys. Res. Lett.*, **29**, p.30, 2002.
- 6- Guillot and Guissani, *J. Chem. Phys.*, **105**, p.255, 1996.
- 7- Wulf et al., *Am. Mineralogist*, **84**, p.1471, 1999.
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