EUROPEAN SYNCHROTRON RADIATION FACILITY

INSTALLATION EUROPEENNE DE RAYONNEMENT SYNCHROTRON

	Experiment title: Compounds of Fe and H under planetary pressures	**Experiment number**: ES1247
Beamline: ID24	**Date of experiment**: from: 11/07/2023 to: 17/07/2023	**Date of report**: 31/08/2023
Shifts: 18	**Local contact(s)**: O. Mathon	*Received at ESRF:*

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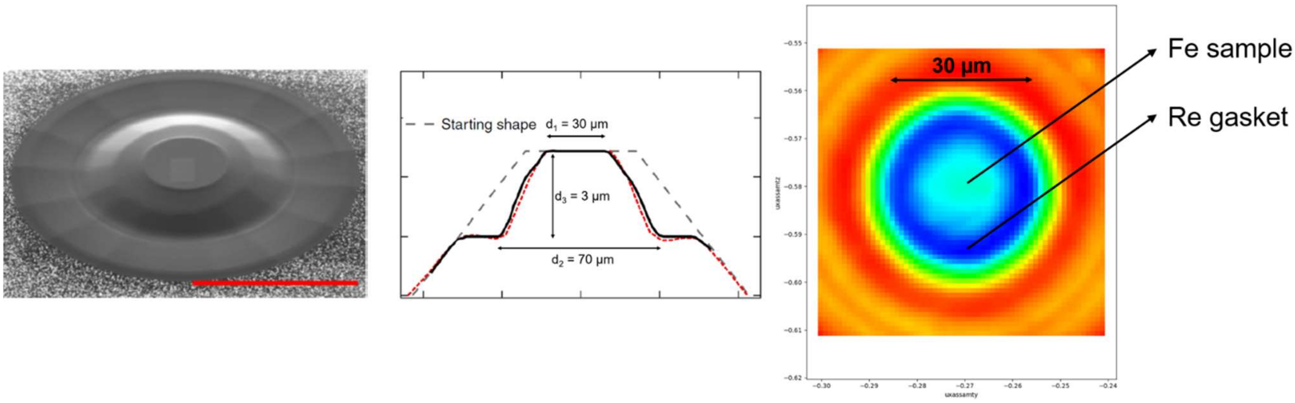
Report:

Fe and H are two major constituents of planetary interiors. It was shown that pressure dramatically increases the H uptake in Fe. Many stoichiometric compounds have been synthesized in the Fe-H system under pressure, from interstitial hydrides, FeH and FeH2 to compounds, FeH3 and FeH5. Yet, the exploration have been limited to 130 GPa. We proposed to explore by combined XAS/XRD experiment the Fe-H phase diagram up to Earth's core conditions, in order to uncover and characterize novel high stoichiometry iron hydrides.

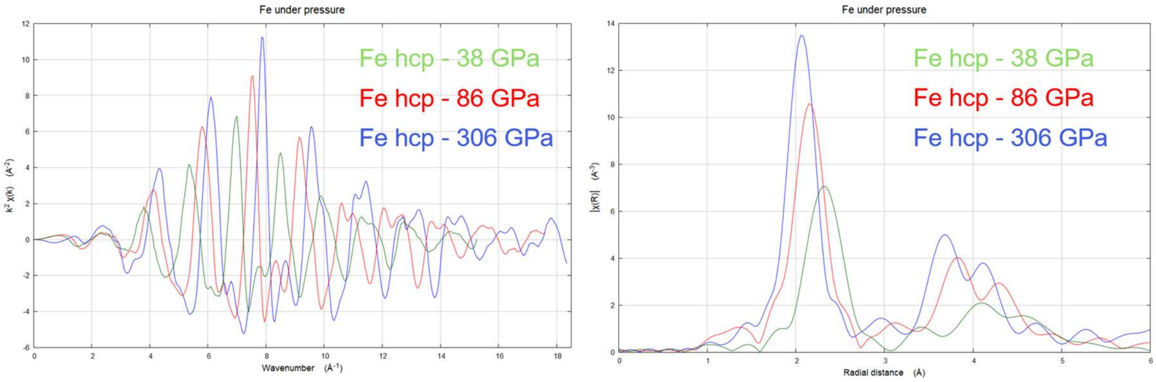
Behind these scientific goals, the instrumental technique towards ultra-high pressure and hydrogen loading represents a challenge by itself, and experience gained during this experiment will be of benefit to the entire high-pressure community. In order to decouple technical challenges, two high pressure cells were loaded. The two cells were prepared with toroidal DAC design [1] using nano-polychrystalline diamonds [2]. Toroidal shapes have been prepared at the CEA, Bruyeres-le-Chatel in France using femto-laser technique. The first cell (cell1) was loaded with a 8 µm thick pure Fe foil from Goodfellow and no pressure transmitting medium. The second cell (cell2) was loaded with a 8 µm thick pure Fe foil from Goodfellow and with NH_3BH_3 salt as pressure medium and source of hydrogen.



Left panel: picture of toroidal diamond. Central panel: characteristics of the toroidal shape used for this experiment. Right panel – Map of the cell performed on the beamline in transmission mode.

<u>First run with cell1</u>

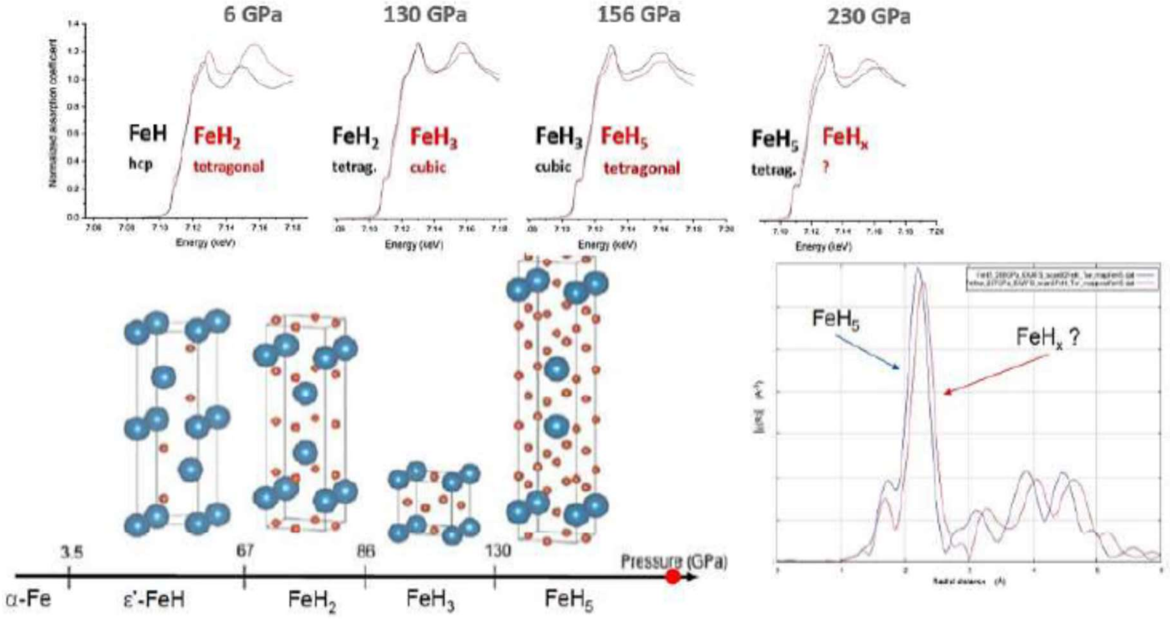
The main goal was to measure the data quality obtained on the new ID24-DCM beamline at ultra-high pressure and to evaluate the mechanical behaviour of toroidal anvils made of NPD, independently of problems induced by hydrogen loading. We reached during this first run pressure above 300 GPa, with excellent signal-to-noise-ratio. EXAFS data allows to determine the first neighbours distance of the hcp structure and determine the pressure in the cell.



Fe EXAFS data – Left panel $K^2.\chi(K)$, right panel FT.

<u>Second run with cell2</u>

The first goal of this run was to reproduce the FeH_x sequence under pressure obtained by Pepin et al. [3]. We successfully reproduce the $Fe/FeH/FeH_2/FeH_3/FeH_5$ sequence, following the pressure/temperature protocol described in [3] (increase of pressure and laser heating of the cell at each pressure point, except for FeH who transforms spontaneously at 6 GPa). For FeH_3 and FeH_5 phases, confirmation of structure and pressure was obtained by combined XRD measurements at the High Pressure XRD beamline ID15B. Well above 130 GPa, another structure appears characterized by an increase of the Fe-Fe distance compared to FeH_5 structure (despite pressure increase) and a modification of the XANES signal. These modifications probably indicates a further evolution of the FeHx structure towards higher stoichiometry with $x \geq 6$. The structure and the pressure have been characterized once again by combined XRD measurement at ID15B. XRD data confirms the evolution of the structure. Recent theoretical papers (for example Kvashnin et al. [4]) predicts stable structure with different H stoichiometry as P4/mmm-Fe_3H_5, Immm-Fe_3H_{13}, I_4/mmm-FeH_5 and Cmmm-FeH_6 phases above 150 GPa. Now, a careful analysis of XANES/EXAFS and XRD data must be performed to determine the new synthetized structure. A very rich and unexplored chemistry seems to appear above 150 GPa.



Top FeH_x XANES data corresponding to the sequence structure proposed by [3] (bottom left figure). Bottom right, EXAFS data above 130 GPa demonstrates an increase of the Fe-Fe distance compare to FeH_5 structure despite the pressure increase, indicating a higher H stoichiometry.

References

[1] Dewael *et al.*, Nat. Comm. **9**, 2913 (2018)
[2] Irifune *et al.*, Nature **421**, 599 (2003)
[3] Pepin *et al.*, Science **357**, 382 (2017)
[4] Kvashnin *et al.*, J. Phys. Chem. C **122**, 4731 (2018)