ESRF	**Experiment title:** **Synthesis of novel $YFe_2H_{(x>10)}$ compounds up to 100 GPa: a possibility to recover a high content H-storage material at ambient pressure**	**Experiment number**: HC5070
Beamline: ID15B	**Date of experiment**: from: 15/11/2022 to:18/11/2022	**Date of report**: 8/12/2022
Shifts: 9	**Local contact(s)**: Gaston Garbarino	*Received at ESRF:*

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Report:

Scientific background and objectives:

Under pressures of few tens of GPa, many elements have been predicted to form poly-hydrides with unconventionally high hydrogen stoichiometry. Interestingly, many of these superhydrides are predicted to have a conventional superconductivity, some with a remarkable almost ambient critical temperature [1]. A surge of ab-initio calculations has guided many experimental studies using synchrotron XRD and conductivity measurements. In particular, FeH_5 was the first super-hydride synthesized about 130 GPa and the observed sequence FeH, FeH_2, FeH_3 and FeH_5 under pressure clearly showed the pressure-induced change of nature in hydrides from interstitial to reorganized [2]. A remarkably high superconductivity temperature of 243 K was measured in YH_6 about 200 GPa [3]. It is now crucial to explore possible routes to recover at ambient pressure such remarkable properties of these rich H compounds. The route of ternary hydrides has been suggested [4].

The experimental study of pressure-induced hydrides in FeH_2 alloy seems a good candidate to demonstrate indeed the validity of the ternary hydride hypothesis. YFe_2 has been observed to readily absorb hydrogen with almost continuous amount, leading to YFe_2H_5 above 0.1 GPa. Interestingly, YFe_2H_5 has been shown metastable at ambient pressure [5]. We have performed DFT calculations which showed that YFe_2H_7 and YFe_2H_{10} should be synthesized by compressing YFe_2 in excess hydrogen in the 25 -50 GPa range. The H atoms go in the interstitial sites of the cubic YFe_2 lattice and induce a distortion of the lattice, from tetragonal to monoclinic. At a given pressure, the volume of the formula unit as a function of hydrogen content is calculated linear, with a slope interpreted as the excess volume of a hydrogen atom in the unit cell. The excess volume has been used previously in our XRD studies to determine the stoichiometry of hydrides under pressure [6]. Finally, it was calculated that YFe_2H_{10} remains near the convex hull at ambient pressure, with a negative formation enthalpy (-17 meV/f.u.) suggesting that YFe_2H_{10} should remain metastable at ambient pressure upon pressure release. If this is verified, the reachable hydrogen storage density would be in the range of 200 KgH_2/m^3 (around 2.8 times the hydrogen liquid density), well within the target for mobile applications.

Another aim is to go in the 100 GPa range to increase further the H content in $YFe_2H_{x>10}$ and observe the transition from interstitial to reorganized hydrides. The two end-member components, Fe and Y, already formed very intriguing super-hydrides, namely FeH_5 and YH_6. The H sub-lattice in $YFe_2H_{>11}$ superhydride might be very interesting. DFT calculations are under progress to guide our experimental search.

The goal of our experiment was the following: 1) use standard DACs to synthesize $YFe2H_{(6\ -\ 10)}$ by compressing $YFe_2H_{4.2}$ in excess hydrogen up to the 80 GPa range. Perform XRD determination of the structure, volume and hence observe the highest stoichiometry stable at a given pressure. Measure the volume upon decompression to show ambient pressure metastability. 2) Synthesize super-hydride of YFe_2H_x by going above 100 GPa.

Experimental details and results

Three diamond anvil cells were prepared covering various pressure range. $YFe_2H_{4.2}$ was loaded with an excess of hydrogen in the four DACs.

A first cell mounted with diamonds with culets of 300µm was loaded with two pieces of $YFe_2H_{4.2}$. XRD measurements were performed up to 51 GPa, leading to an equation of state for YFe_2H_x (see fig. 1). During compression, we notably observe a jump of stoichiometry from x(H)=4.5 at 16 GPa to x(H)>6.5 at 18 GPa. The hydrogen stoichiometry stays constant up to 51 GPa. This validates the previously predicted formation of YFe_2H_7 near 20 GPa. However, it invalidates the previously predicted formation of YFe_2H_{10} near 20 GPa.

We investigated the metastability of YFe_2H_7 with another cell by decompressing it while taking XRD measurements. The decompression started at 30 GPa (see fig. 1). First, it appears that the hydride continuously loses hydrogen atoms from 20 GPa to 10 GPa, then a jump of hydrogen stoichiometry occurs at lower pressure, from x(H)=6 at 3 GPa to x(H)=5.3 at 1 GPa. Thus, we did not find that YFe_2H_7 was metastable.

Another DAC, with culets of 150µm was loaded with $YFe_2H_{4.2}$, in order to investigate the formation of YFe_2H_{10} at a higher pressure range, up to 90 GPa. XRD measurements were made but the results need more analysis. At first glance, there seems to be a mix of different phases.

To conclude, in the present experiment we managed to form YFe_2H_7, to obtain complete structural data by measuring the single-crystal diffraction of this hydride, and to show that YFe_2H_7 is not metastable under ambient pressure. We now need to further explore the megabar pressure range in order to carefully investigate the formation of YFe_2H_{10}. A new proposal will be submitted to pursue this study.

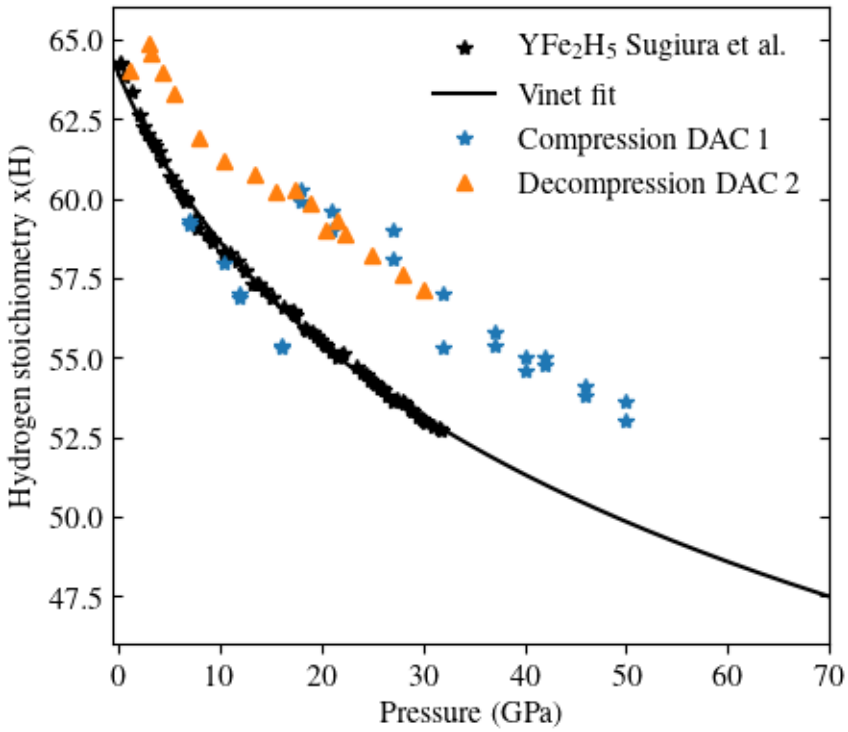

Figure 1: Equations of state during compression and decompression of YFe_2 hydride, and equation of state of YFe_2H_5 extracted from reference [7].

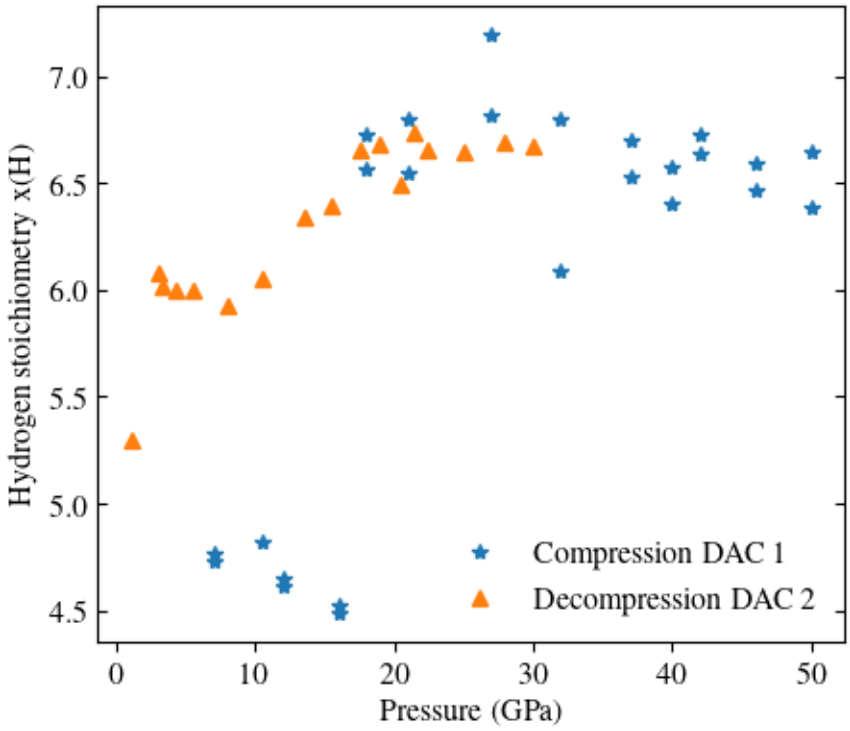

Figure 2: Estimated stoichiometry under pressure during compression and decompression. The stoichiometry was obtained by the volume expansion per YFe2 formula unit [7].

References

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