

**HYDROGEN ISOTOPIC RATIO IN IRON METEORITES.** C. Defouilloy<sup>1</sup>, R. Duhamel<sup>1</sup>, F. Robert<sup>1</sup>, M. Clog<sup>2</sup>,  
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**Introduction:** Iron Meteorite are generally assumed to come from differentiated planetesimals accreted very early in the chronology of the Solar System. Various radiogenic chronometers show that the core formation (Hf-W) of these parent bodies occurred 1,5 My after de CAI condensation [1][2], and that the metal crystallization (Pd-Ag, Mn-Cr, Re-Os) took place within 10 My [1].

This scenario could be better documented via the determination of the isotopic compositions of the Hydrogen contained in Iron Meteorites.

The D/H ratio being significantly different between the protosolar Hydrogen ( $(D/H)_{H_2} = 20 \pm 5 \times 10^{-6}$  [3]) and Hydrogen coming from water reduction during planetesimal differentiation ( $(D/H)_{\text{condritic water}} = 150 \times 10^{-6}$  [3]) it can be used as an indicator of the origin of Hydrogen in the Iron Meteorites. Therefore, an analytical technique for measuring the D/H ratio of Iron meteorites using ion microprobe has been developed [4].

**Method:** Hydrogen isotopic composition is determined with the CAMECA IMS 3f at the National Museum of Natural History in Paris, France. Samples are mounted either in epoxy and Au-coated or as polished thin sections and sputtered with a primary positive  $Cs^+$  beam. Because of the highly conductive nature of these metallic samples, the use of an electron gun is unnecessary. Entrance and Exit slits as well as the Energy slit are kept wide open. Under these conditions, the mass resolution is  $<400$  (with the  $Cs^+$  primary beam the possible mass interference between  $H_2^-$  and  $D^-$  is negligible). Moisture from evaporation of absorbed water in the ion probe was kept as low as possible by using a liquid nitrogen trap.

Experiments showed that with a primary beam intensity ranging from 5 to 10 nA with a 30  $\mu m$  diameter beam, it is impossible to eliminate efficiently all the contribution of absorbed surface water. Therefore, we used a larger ion spot (500  $\mu m$ ) combined with a 150  $\mu m$  aperture field, so that the ions from the center of the "crater" are selectively collected. The intensity had to be increased up to 40 nA in order to reach at least  $10^4$  cps for H and a few cps for D. Data were collected in peak jumping mode, with counting time of 5 and 15 s for  $H^-$  and  $D^-$  respectively.

We created our own standard sample by measuring the D/H ratio of a sample we rusted with distilled water with a known D/H ratio. In a first approximation we assumed that the reduction of water by the metal was

negligible. The instrumental fractionation factor is then of  $0.58 \pm 0.04$ .

We also implanted a known Hydrogen quantity into Golden foils in order to calibrate the Hydrogen concentrations of our sample in an absolute way. This calibration showed that roughly 1% of the implanted Hydrogen atoms is detected when analysed with the 3f microprobe.

**Results:** The D/H ratio has been investigated in 8 Iron meteorites (Copiapo IAB, Toluca IA, Coahuila IIA, Cape of Good Hope IVB, Ballinoo IIC, Perryville IIC, Mont Dieu UnGr, Tucson UnGr), as well as Ben-cubbin and Disko Island, a terrestrial iron deposit. Results are obtained as depth profiles of H and D. figure 1 shows the reproductibility of 5 depth profiles obtained for the Mont Dieu meteorite.

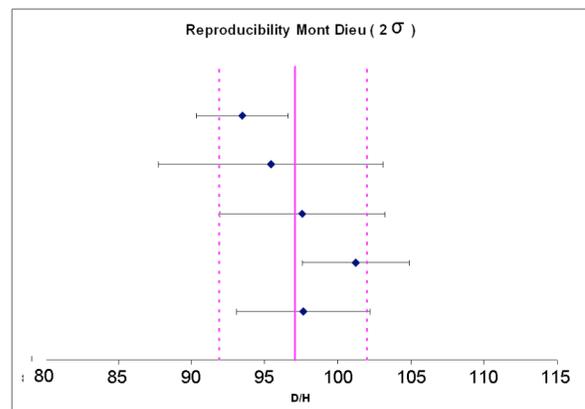


Fig. 1. Reproducibility diagram of the D/H ratio measured in Mont Dieu. The mean D/H value is of  $97 \pm 5 \times 10^{-6}$  ( $2\sigma$ ); ie  $\delta D = -409 \pm 32 \text{ ‰}$ .

Our main concern was to assess any possible source of pollution. This pollution could have 3 main sources : alcohol during sewing and polishing, development of rust at the surface of the sample, and exchange with the atmospheric moisture.

The polishing of a sample with a highly deuterated alcohol ( $D/H = 10^{-2}$ ) showed that this possible contamination was negligible [4].

Several experiments were conducted to determine the influence of rust. Samples from Copiapo and Mont Dieu were put in contact with water. Results show that the rust layer is confined to the surface and clearly identifiable on a O/Fe vs. H/Fe diagram (figure 2), but

the diffusion of the water hydrogen goes deeper into the sample.

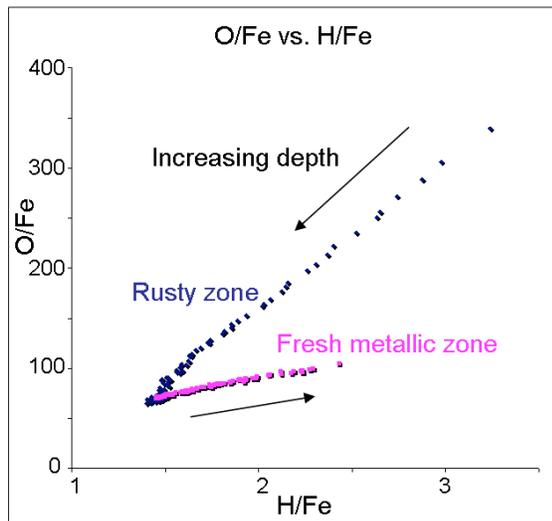


Fig 2.  $O/Fe$  vs.  $H/Fe$  diagram realized on the superficially rusty sample from Mont Dieu. The rusty zone at the surface of the sample is clearly visible.

Finally, we tried to quantify the contamination by the atmospheric moisture. To do so, we conducted a series of experiments with different D/H ratios (from 200 to 600  $10^{-6}$ ) for water in contact with samples from Mont Dieu. These experiments were short enough not to let rust forms at the surface. We calculated a contamination of  $47 \pm 21\%$ . Thus the indigenous D/H ratio of Mont Dieu is lowered to  $67 \pm 30 \times 10^{-6}$  (figure 3).

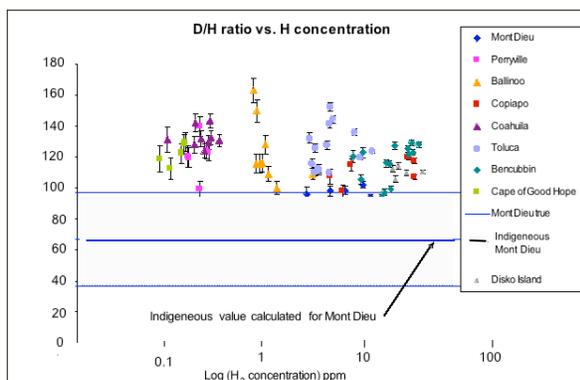


Fig 3.  $D/H$  ratios measured in 7 meteorites, as well as Bencubbin, and the terrestrial Disko Island. Correction from a possible atmospheric pollution calculated for the Mont Dieu meteorite lowers the ratio to an area around  $70 \times 10^{-6}$  (Indigenous value).

**Discussion:** In order to interpret our data, we also need to take into account the Deuterium created by spallation during the exposure time of our samples. Some samples contain traces of Hydrogen - and Deuterium... The calculated H concentration can be as low as 0.1 ppm for Cape of Good Hope, Coahuila and Perryville (figure 3).

We failed to obtain a correct measure of the D/H ratio for Tucson, which does not contain enough Hydrogen to make a satisfactory D/H ratio. The H count rate was around  $10^3$  cps, which is insufficient to measure a stable D count rate. Thus, Tucson was not included in our study.

Assuming that the spallation rate is the same for all our samples, we considered that all the deuterium from the less concentrated (but measurable) sample, Cape of Good Hope, was spallagenic to calculate a maximum value of the Deuterium created by spallation. This gives us a first approximation of the spallagenic deuterium for all the samples, which appears to be negligible for all of them, apart the 3 containing very little Hydrogen (Cape of Good Hope, Coahuila and Perryville)

In order to complete our study, we now need to determine the carrying phase of Hydrogen. This Hydrogen can be either as -OH, -CH, or dissolved in the metal. Measuring the possible relation between H and O or C should allow us to answer that question.

A Rayleigh distillation of water on Iron performed at the LGIS IPG-Paris, France, gave us an isotopic fractionation factor between water and hydrogen of  $\alpha = 1.49 \pm 0.16$ . This would give, for instance for the Mont Dieu sample, an initial D/H ratio of  $100 \pm 45 \times 10^{-6}$ . This value is in agreement with a possible chondritic water origin for the Hydrogen.

**References:** [1] Scherstén A. et al. (2006) *Earth & Planet. Sci. Letters*, 241, 530-542. [2] Blicher-Toft J. et al. (2010) *Earth & Planet. Sci. Letters*, 296, 469-480. [3] Deloule E. and Robert F. (1995) *Geochim. Cosmochim. Acta*, 59, 4695-4706. [4] Defouilloy C. et al. (2010) LPS XLI, Abstract #1340.