

HYDROGEN ISOTOPIC COMPOSITIONS AND WATER CONTENTS IN TYPE I CHONDRULES OF PARIS CM CHONDRITE. A. Stephant¹, L. Remusat¹ and F. Robert¹, ¹Laboratoire de Minéralogie et Cosmochimie du Muséum, UMR CNRS 7202, Muséum National d'Histoire Naturelle, CP 52, 57 rue Cuvier, 75231 Paris, France. E-mail : astephant@mnhn.fr.

Introduction: Hydrogen in aqueously altered chondrites is carried by organics and hydrated minerals. Chondrites show a systematic enrichment in deuterium relative to the molecular hydrogen of the proto-solar nebula [1]. The origin of this deuterium enrichment in chondrites is a key for understanding parent bodies formation processes and the origin of volatils in the solar system [2].

The whole-rock D/H ratios in chondrites has been assumed to reflect the D/H ratio of water that has circulated during the aqueous alteration of the parent body [3]. However, Deloule et Robert [4,5] performed hydrogen isotopic measurements on Bishunpur, Semarkona LL3 chondrites and Renazzo CR2 chondrite and these chondrites exhibit large variability of D/H ratio in matrix clay minerals and also on chondrule silicate minerals. This implies that another process than global isotopic reequilibration during hydrothermal alteration needs to be considered in order to explain the H isotope distribution in chondrites.

Here we analyzed the water content and the D/H ratio on different phases, olivine, pyroxene and mesostase, of 12 type I chondrules and surrounding matrix from Paris which is considered as a slightly altered CM chondrite (CM3.0±0.1) [6]. In comparison, 4 chondrules of Bishunpur (LL3) and 3 chondrules of Renazzo (CR2.0) were also analyzed.

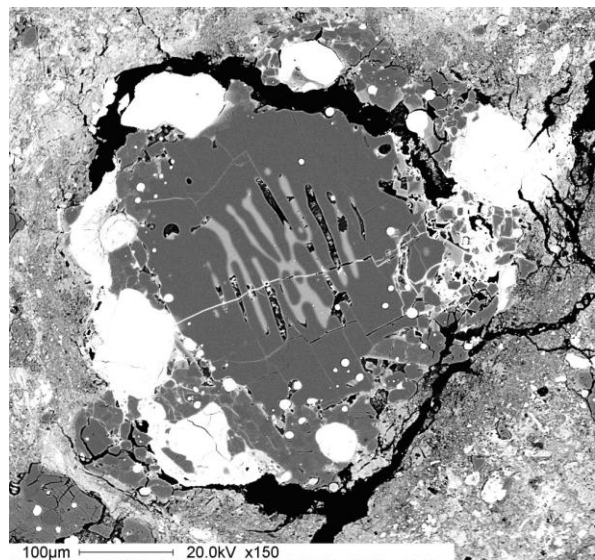


Fig. 1: SEM image of a preserved Type I chondrule in Paris.

Methods: Preliminary characterizations of type I chondrules composition were carried out by SEM (Fig. 1) and electron microprobe on polished sections of Bishunpur, Renazzo and Paris. Two other epoxy-free sections of Paris were also studied.

Elemental and isotopic measurements were done with the Cameca NanoSIMS 50 at the Muséum National d'Histoire Naturelle in Paris. Two steps were necessary for these measurements: on a first step $^{16}\text{OH}^-$, $^{28}\text{Si}^-$, $^{24}\text{Mg}^{16}\text{O}^-$ and $^{27}\text{Al}^{16}\text{O}^-$ were analyzed and on a second step H^- and D^- were analyzed; both with a cesium primary beam. For water content, a primary beam with a current of 15pA and a 200µm aperture diaphragm was rastered on a 20×20µm² surface area divided in 256×256 pixels. The counting time was set to of 1ms/px. Prior to the acquisition, a 25×25µm² surface area was presputtered using a larger current. Special attention was dedicated to hamper the contribution of water contamination. The same procedure was used for isotopic measurements except that a primary beam of 100-240pA for Paris and 29pA for Bishunpur and Renazzo. The same areas were imaged for both elemental and isotopic distributions. 63 images of both elemental composition and isotopic ratio in type I chondrules were obtained for Paris, 22 for Bishunpur and 17 for Renazzo. A set of silicate standards with known compositions was used for calibration and instrumental isotopic mass fractionation correction.

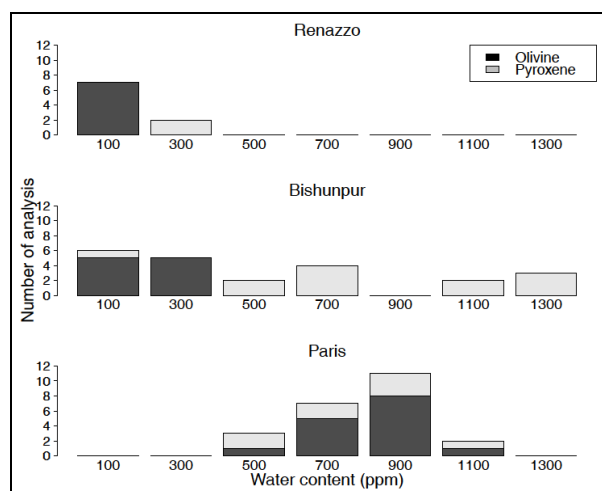


Fig. 2: Water content distribution in olivine and pyroxene chondrule minerals of Renazzo, Bishunpur and Paris chondrites.

Results:

Water content: Calculations were done using OH^-/Si^- versus $\text{H}_2\text{O}(\text{ppm})/\text{SiO}_2(\text{wt}\%)$ calibration. This calibration is a function of the silicate mineralogy [7], and thus, is different for olivines, pyroxenes and mesostases. Distribution of water contents in olivines and pyroxenes of type I chondrules are presented in Figure 2 for each chondrite. Pyroxene mean water contents are $343 \pm 33 \text{ ppm}$, $894 \pm 335 \text{ ppm}$ and $770 \pm 216 \text{ ppm}$ for Renazzo, Bishunpur and Paris respectively. In olivine, the water concentration varies markedly between Renazzo, Bishunpur and Paris: $156 \pm 44 \text{ ppm}$, $212 \pm 127 \text{ ppm}$ and $832 \pm 127 \text{ ppm}$, respectively. Paris chondrules appear to be more hydrated than those from Bishunpur and Renazzo and do contain significant amount of water in nominally anhydrous minerals. In mesostases, water concentration varies in the range of several wt% H_2O .

D/H ratio: Distribution of the hydrogen isotopic composition of minerals in chondrules from Paris, Bishunpur and Renazzo are shown in Figure 3. Renazzo D/H ratio in chondrule olivines, pyroxenes and mesostasis range from 129×10^{-6} to 162×10^{-6} with a mean value of $138 \pm 9 \times 10^{-6}$; Bishunpur range from 133×10^{-6} to 181×10^{-6} with a mean value of $147 \pm 15 \times 10^{-6}$. Paris exhibits a wider distribution from 94×10^{-6} to 213×10^{-6} with a mean value of $142 \pm 22 \times 10^{-6}$. The mean D/H ratios for these three meteorites fall in the terrestrial domain; that is $156 \pm 10 \times 10^{-6}$. In Paris, the D/H ratio of the surrounding matrix presumably standing for the isotopic composition of the alteration water is $145 \pm 5 \times 10^{-6}$. In addition to this broad distribution, we notice that intra-chondrule heterogeneities are also pronounced in Paris. Indeed, in a single chondrule, the isotopic variability can reach 75×10^{-6} among olivines (i.e. 480%).

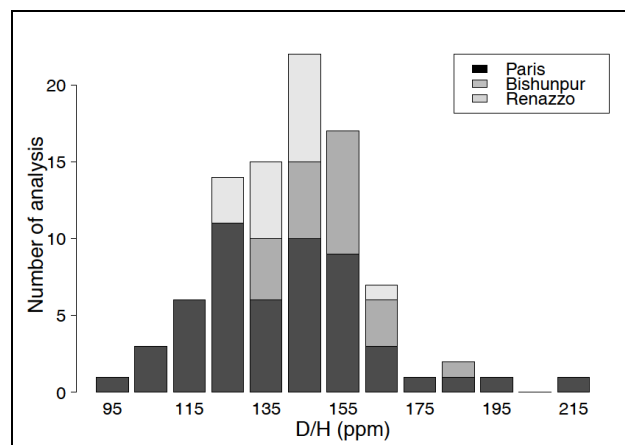


Fig. 3: Distribution of D/H ratio analyzed in chondrules minerals of Paris, Bishunpur and Renazzo.

Discussion: Chondrules in Paris contain water concentration markedly in excess for nominally anhydrous minerals. These minerals have recorded a process that led to a broad distribution in their hydrogen isotopic ratios. In principle, late hydrothermal alteration of the minerals in these chondrules should result in isotopic equilibration and thus variations impeded by isotopic fractionation factors under equilibrium; this implies that the isotopic variations should be restricted at most within 30 ppm (i.e. 200%)

An isotopic exchange between the different minerals of the same chondrule cannot be the cause of the measured variations since, in addition to isotopic heterogeneities among chondrules, intra-chondrule variations reach 109×10^{-6} , which corresponds to 700%! In terrestrial environments, such variations are never observed among alteration products and this is clearly an evidence against a simple equilibrium isotope fractionation between water and chondrules, resulting from an hydration of anhydrous minerals, or between the minerals of the same chondrule.

The possible contamination of the samples by terrestrial moisture can also be ruled out since chondrules have recorded both enrichment and depletion of deuterium relative to the terrestrial value. This study brings new questions on chondrules formation that appear to have recorded a specific and yet unknown process.

References: [1] Robert F. (2002) *MessII*, xx. [2] Alexander C.M.O'D. (2012) *Science*, 337, 721–723. [3] Eiler J. M. & Kitchen N. (2004) *Geochim. Cosmochim. Acta*, 68, 1395–1411. [4] Deloule E. et Robert F. (1995) *Geochim. Cosmochim. Acta*, 59, 4695–4706. [5] Deloule E. et al. (1998) *Geochim. Cosmochim. Acta*, 62, 3367–3378. [6] Bourot-Denise M. et al. (2010) *LPS XLI*, Abstract #1683. [7] Tenner T. J. et al. (2009) *Chem. Geol.*, 262, 42–56.