

ORIGIN OF WATER IN METEORITES : ION-PROBE DETERMINATIONS OF D/H RATIOS IN CHONDRULES.

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Water contents and H isotopic compositions have been determined with the CRPG-Nancy ion microprobe in twelve chondrules (type I and type II after McSween's classification) from several deuterium-rich LL3 meteorites. The water concentration ranges from 500 to 17,000 ppm, with most of the data lying between 1000 and 5000 ppm and the δD_{SMOW} values between -510‰ and +1930‰ (*i.e.* D/H ratios between 76 and 457 x 10⁻⁶). Several spots were analysed in each chondrule to test their internal homogeneity. In an individual chondrule, the distribution of both water concentration and its isotopic composition could be either quite homogeneous, either extremely heterogeneous, but no systematic gradients were found. These ion microprobe measurements demonstrate that hydrogen bearing components (water and organics) were present in chondrule precursors.

I. Introduction

Water contents (expressed as H₂O wt. %) and their corresponding H isotopic compositions have been determined with the CRPG-Nancy ion microprobe in twelve chondrules from several deuterium-rich LL3 meteorites. These chondrites are known to exhibit - as a whole rock - a marked enrichment in their D/H ratios relative to terrestrial values. They are therefore ideal samples to address the problem of the origin of hydrogen in chondrules since terrestrial contamination clearly has a different isotopic signatures (with D/H ratios around 120-160 x 10⁻⁶) from those of indigenous phases (D/H up to 450 x 10⁻⁶).

Results of isotopic analyses obtained on individual chondrules have been reported in the literature (Robert *et al.*, 1979; Robert *et al.* 1987; Sears *et al.*, 1995). In some cases it has been shown that hydrogen with non-terrestrial D/H ratios, was outgassed from chondrules (same refs.). However, no firm conclusions can be derived from such pyrolysis experiments, because LL3 matrix is known to contain compounds (organic macromolecules and water) with extremely high D/H ratios, probably of interstellar origin. Therefore, matrix contamination at the surface of the chondrules or matrix inclusions embedded into the chondrules, are possible sources for the high D/H ratios found in chondrules. In our analyses, all the data have been acquired from well-identified olivine or pyroxene crystals and in the glass. Therefore the current data truly represent hydrogen dissolved within the main mineralogical constituents of the chondrules.

II. Experimental

After calibration on terrestrial olivines and glasses, the relative precision in water concentration is estimated to lie between ± 50 and $\pm 7\%$ for concentrations ranging between 500 to 10,000 ppm, respectively. The precision on the D/H ratios is $\leq \pm 10\%$ for water concentrations higher than 1,000 ppm. The analysed chondrules belong to type I and type II (after McSween's classification; type I stands for highly reduced olivine *i.e.* Mg/Fe+Mg close to unity while type II have Mg/Fe+Mg ≤ 0.85). The range of water concentration is 500 to 17,000

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ppm, although most of the data lie between 1000 and 5000 ppm. The δD_{SMOW} values range between -510‰ and $+1930\text{‰}$ (*i.e.* D/H ratios between 76 and 457×10^{-6}). This complete variation is observed between different chondrules from a single polished section of the Bishampur meteorite. Three to ten spots were analysed in each chondrule in order to test their internal homogeneity in concentration and in isotopic composition. In an individual chondrule, the distribution of both water concentration and its isotopic composition could be either quite homogeneous, either extremely heterogeneous. For example, in chondrule Ch.8, 7 spots exhibit δD values between -370‰ and $+1540\text{‰}$ (*i.e.* D/H ratios between 58 and 395×10^{-6}) while in chondrule Ch.9 they are restricted between -63‰ and -5‰ (*i.e.* D/H ratios between 146 and 155×10^{-6}) for 3 spots. No systematic gradients were found in these profiles. Interestingly two chondrules systematically exhibit δD values lower than those unusually associated with the terrestrial environment (down to -510‰). A similar observation was reported for two sets of chondrules pyrolysed under helium (Robert *et al.* 1987) and therefore there is little doubt that a deuterium-depleted component exists in the ferro-magnesian silicates of the chondrites.

These isotopic data demonstrate that water in chondrules results neither from terrestrial contamination nor by diffusion of water into the chondrule during the hydrothermal alteration in the parent body, since both processes would have produced a homogeneous isotopic composition within each chondrule and within each meteorite. In addition, since the two end members of the D/H distribution (D/H from 58 to 456×10^{-6}) are typically extraterrestrial, the intermediate terrestrial like ratios, result from the mixing of these two end members and not from terrestrial contamination. This is a central conclusion as far as the origin of water in the solar system is concerned. In fact D/H ratios below 60×10^{-6} represent the least fractionated hydrogen component compare to the protosolar H_2 ($20 \pm 10 \times 10^{-6}$).

III. Implications

These ion microprobe measurements demonstrate that hydrogen bearing components (water and organics) were present in chondrule precursors. The sources for these components are 1) the interstellar medium for the deuterium rich phases (with $\text{D}/\text{H} > 150 \times 10^{-6}$; Deloule & Robert, 1995) and 2) a local oxidation of the protosolar nebula hydrogen resulting in the formation of water-bearing minerals (Lécluse and Robert, 1994). The large differences between the isotopic composition of these phases were not homogenised during the flash heating and the rapid cooling of the chondrules. As a consequence, chondrules represent a potential water reservoir in the primitive planets.

References

Robert *et al.*, *Nature* **282**, 785-789 (1979). Robert *et al.*, *Geochim. Cosmochim. Acta* **51**, 1787-1805 (1987). Lécluse & Robert, *Geochim. Cosmochim. Acta* **58**, 2927-2939 (1994). Sears *et al.*, *Meteoritics*, **30**, 169-181 (1995). Deloule & Robert, *Geochim. Cosmochim. Acta* **59**, (1995).