HYDROGEN ABUNDANCES IN METAL GRAINS FROM THE HAMMADAH AL HAMRA (HaH) 237 METAL-RICH CHONDRITE: A TEST OF THE NEBULAR-FORMATION THEORY.

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Introduction: The Bencubbin-like (CB) chondrites are metal-rich, primitive meteorites [1,2]. Some of these chondrites (HaH 237, QUE 94411) contain compositionally zoned metal grains with near-chondritic bulk compositions. Thermodynamic modeling of the zoning patterns in these grains suggests that they were formed by condensation in a region of the solar nebula with enhanced dust/gas ratios and a total pressure of 10^{-4} bars at temperatures between 1400 – 1500 K [3]. If these predictions are correct than the metal grains would have been exposed to abundant H_2 gas, which comprises the bulk of nebular systems. Since Fe-based alloys can absorb significant quantities of H, metal grains formed in the solar nebula should contain measurable abundances of H.

Theory: The ability of metals to absorb hydrogen has been known since the mid 1800s [4]. The properties of metal-hydrogen systems have been the subject of intense research because small concentrations of H in metal results in significant changes in mechanical and metallurgical properties, particularly the brittleness of stainless steels and other industrial metals [5]. Within a metal, hydrogen molecules are dissociated and hydrogen atoms occupy interstitial sites in the host-metal lattice. Hydrogen atoms jump from one interstitial site to a neighboring vacant one and diffuse this way over large distances through the metal, resulting in rapid H saturation in a metal grain. Furthermore, defects in metallic microstructures act as hydrogen traps that significantly increase the solubility of H and decrease its mobility [5]. Once in these defects sites, it is very difficult to remove H from metal; a major problem for the steel industry but a potential advantage for cosmochemistry.

The H concentration in metal (X_H) is given by [6]:

$$X_H = \sqrt{P_{H_2}} \cdot e^{-\frac{\Delta S^{nc}}{k_b}} \cdot e^{\frac{\Delta H}{k_b T}} \qquad (1)$$

In equation (1) P_{H2} is the partial pressure of H_2, ΔS_{nc} is the non-configurational entropy of solution, k_b is Boltzmann's constant, ΔH is the enthalpy of solution, and T is temperature. Values for ΔS_{nc} and ΔH for metallic Fe and Ni are tabulated in [7].

Using equation (1), we calculated the abundance of H in Fe metal in contact with a solar nebula gas. If metal grains condensed from an ambient solar composition gas at 10^{-4} bars total pressure at a temperature of ~1450 K, then they should have incorporated ~3 ppm H. Thus, a direct test of nebular formation of zoned metal grains is to measure the abundance of H in metal grains that are purported nebular condensates. Such measurements have the potential to not only establish whether metal formed in the presence of H_2 but also constrain the partial pressure of this species.

Analysis: A polished slab of the metal-rich chondrite HaH 237 was analyzed. Elemental distributions of Mg, Si, P, and Ni were obtained via wavelength dispersive X-ray mapping using the Cameca SX-50 electron microprobe at U of A. Using these data both a metal grain with significant zonation in Ni and a relatively homogeneous unzoned metal grain were selected for detailed analysis. The concentrations of Si, P, S, Cr, Fe, Ni, Co, and Cu were determined in traverses across these grains using electron microprobe analysis.

Hydrogen analysis was carried out using the Cameca IMS 6f ion microprobe at ASU. Standard Reference Material (SRM) standards and a polished thick section of HaH 237 were mounted in epoxy-free disks. To further reduce H background in the ion microprobe, the probe was baked for over 24 hours prior to the analytical session. Data were collected when the ultrahigh vacuum reading of the sample chamber was at ~$2.3\text{-}5\times10^{-10}$ torr. A 10 kV Cs^+ beam of ~2 nA, tuned into a spot of ~25 μm in diameter in aperture illumination mode, was used to sputter samples. Secondary ions were accelerated to -9 kV and collected through a small field aperture at low mass resolution conditions. Fe was used as the reference element for hydrogen quantification.

The Standard Reference Material NBS 1095 AISI 4340 steel was used as our standard. This steel is ~96 wt.% Fe, 1.8 wt.% Ni, 0.8 wt.% Cr, and 0.7 wt.% Mn. It is certified to contain <5 ppm H. Ion images indicated that H in the SRM and HaH 237 metal is homogeneous over each spot analyzed.

Results: The zoned metal grains contain variable Ni abundances with ~5 wt.% Ni at their rim and 12 wt.% in their core (Figs. 1 – 2). The unzoned metal grain contains ~6.5 wt.% Ni throughout most of its interior with the exception of a Ni-rich area containing >8 wt.% Ni near the core (Figs. 3 – 4).

Both metal grains in HaH 237 contain abundant H. The limit of H in these grains is ~30 times that in the standard. We use an extremely conservative estimate of 1 ppm for the H in the SRM to constrain the H abundance in the chondritic metals. The zoned metal grain contains a minimum of 37 ppm H at its core and 29 ppm at the rim. Averaging all analyses suggests that the zoned metal grain contains >28 ± 9 ppm H. Analyses of the unzoned grain yields >28 ± 5 ppm H.

Discussion: The observed H abundances determined for metal grains in HaH 237 are significantly higher than those calculated using equation (1) for the conditions determined from thermodynamic modeling [3]. There are two possibilities to explain this discrepancy: 1) the metal grains condensed under a significantly higher H_2 partial pressure or 2) the minor elements in metal increase its H absorbing properties.

If we assume scenario 1), then we can constrain the P_{H2} during metal condensation. A metal grain in contact with H_2 gas at 1450 K will incorporate 8 ppm H at a P_{H2} of 10^{-3} bars and 26 ppm at 10^{-2} bars. Thus, the observed H abundances are consistent with metal condensation in a pressure-enhanced region of the solar nebula. A 100-fold pressure increase is predicted by shock wave models for chondrule formation [8,9].

Our calculations are for pure, perfect metallic crystals of Fe. Similar calculations for pure Ni yield 7 ppm H at 10^{-4} bars, 22 ppm at 10^{-3} bars, and 69 ppm at 10^{-2} bars. Thus, the presence of Ni in the alloy will likely significantly increase the metals ability to absorb H. Furthermore, the defect concentration will increase with the addition of minor and trace elements. These complications make it more difficult to constrain the P_{H2} recorded by the metal grains. However, the presence of abundant H in these grains strongly supports the idea that metal grains in the CB chondrites formed in the solar nebula.

References: [1] Weisberg et al (2001) *MAPS* **36**, 401. [2] Krot A. N. et al. (2002) *MAPS* **37**, 1451. [3] Petaev et al. (2001) *MAPS* **36**, 93. [4] Graham (1866) *Phil. Trans. Roy. Soc. (London)* **156**, 399. [5] Dos Santos and De Miranda (1997) *J. Mat. Sci.* **32**, 6311. [6] Wipf (2001) *Physica Scripta* **T94**, 43. [7] Fukai (1993) *The Metal-Hydrogen System*, Springer-Verlag. [8] Desch and Connolly (2002) *MAPS* **37**, 183. [9] Ciesla and Hood (2002) *Icarus* **158**, 281.

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Fig.1

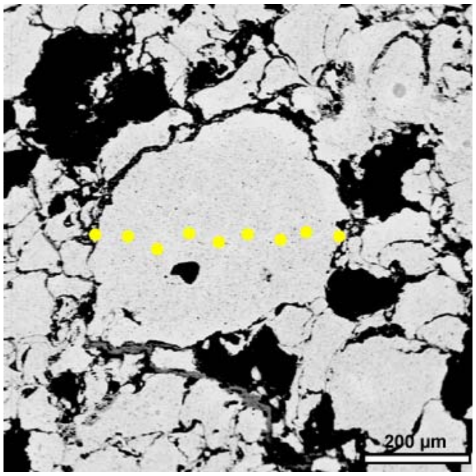

Fig.2

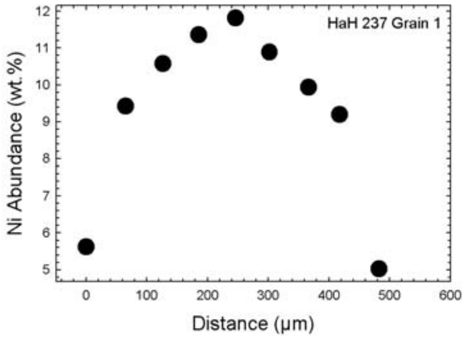

Fig.3

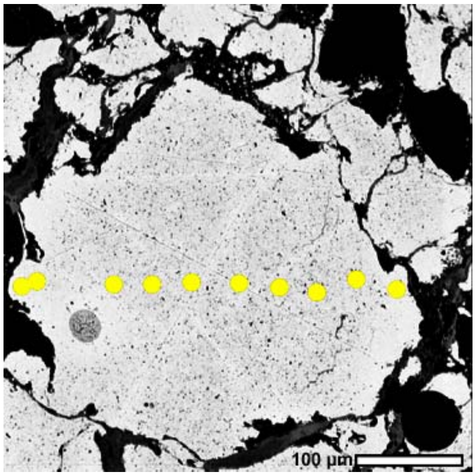

Fig.4