A HENRY'S LAW TEST FOR EXPERIMENTAL PARTITIONING STUDIES OF IRON METEORITES.

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Introduction: Magmatic iron meteorites are believed to be samples of the metallic cores of asteroid-sized parent bodies. Elemental trends within each iron meteorite group are thought to result from fractional crystallization of once completely molten metallic cores [1]. Experimentally determined solid metal/liquid metal partition coefficients have been used to model the crystallization processes inferred to occur within asteroidal cores [2-6].

However, concern regarding Henry's Law has been raised about applying the experimentally determined partition coefficients to the crystallization of iron meteorites [7]. Most solid metal/liquid metal experiments have been analyzed by the electron microprobe, which, because of the detection limits of the instrument, commonly requires experimental samples to be doped with the elements of interest near wt% levels. Such doping levels are often much higher than the natural elemental concentrations measured in iron meteorites.

Further, for other commonly measured elements in iron meteorites, such as As, Cr, and Cu, few experimental solid metal/liquid metal partitioning data exist at all. The concentration of light elements such as S and P in the metallic liquid can significantly affect the solid metal/liquid metal partitioning behavior [2,3]. Understanding such effects on the partition coefficients is necessary to model the behavior of the elements during the crystallization process.

We have experimentally determined new solid metal/liquid metal partition coefficients for P, Ni, and 11 trace elements relevant to iron meteorites using laser ablation ICP-MS microanalysis [8,9]. Because of the analytical sensitivity, we were able to dope our samples at close to natural abundance levels, including trace element concentrations comparable to those observed in magmatic iron meteorites [e.g., 7]. By comparing our low-level doped runs to previous experimental results, we tested Henry's Law.

Experimental and Analytical Methods: Mixtures of dominantly Fe, Ni, FeS, and P were used as starting materials. Eleven trace elements were measured: Ag, As, Co, Cr, Cu, Ga, Ir, Os, Pd, Re, and W. Trace elements were added at a range of concentrations which varied between experiments and elements. The highest doping concentrations were wt% levels while the lowest doping levels were ppb.

Starting mixtures were contained in alumina crucibles and sealed in evacuated silica tubes. The silica tubes were held in a Deltech vertical tube furnace at 1050-1250°C for 4-19 days, depending on temperature. Run products contained homogenous solid metal with about 10 wt% Ni. Run products also contained quenched metallic liquid with a texture of Fe-Ni dendrites surrounded by interstitial P-rich and S-rich phases. Because the starting material contained both P and S, some experiments produced two immiscible metallic liquids. The major element compositions of two runs that contained immiscible metallic liquids is detailed in [10] along with images of the samples.

Trace and major elements were analyzed using laser ablation ICP-MS. Laser ablation ICP-MS analyses were performed using a CETAC LSX-200 laser ablation peripheral with a magnetic sector ICP mass spectrometer, the Finnigan Element™ [8,9]. The solid metal in each run was analyzed with point analyses 150 μm in diameter and 10 μm deep. To minimize the effects of the quench texture, each region of quenched metallic liquid was analyzed by ablating line scans 150 μm wide, 15-30 μm deep and 500-1000 μm long. The isotopes ^{31}P, ^{34}S, ^{53}Cr, ^{57}Fe, ^{59}Co, ^{60}Ni, ^{63}Cu, ^{69}Ga, ^{75}As, ^{105}Pd, ^{107}Ag, ^{182}W, ^{185}Re, ^{192}Os, and ^{193}Ir were monitored during each laser ablation ICP-MS analysis. Instrumental sensitivity factors for each siderophile element were determined by measuring signal intensity from standards (Hoba IVB, NIST SRMs 1263a and 1158) having known concentrations of the elements of interest [8,9]. Analytical precisions for each element were calculated as the standard errors of five replicate analyses of each phase in each run product.

Major element compositions of the phases were also determined by electron microprobe, using a defocused 50 μm beam on the quenched metallic liquid [10]. Compositions determined by the two analytical techniques were consistent and averaged together.

Results and Implications: Figure 1 shows our partitioning results for three trace elements, As, Ga, and Ir, frequently discussed in iron meteorites. The molar solid metal/liquid metal partition coefficient, k, is plotted as a function of the molar concentration of P and S in the metallic liquid, X_P and X_S respectively. The formulation is from [11], who using thermodynamic considerations suggested that such a parameterization should produce linear trends, as seen in Fig. 1. Our experiments contained both S and P while previous experiments [2,3,11-15] shown in Fig. 1 were

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either S-bearing or P-bearing runs. It should be noted that at high S and P contents, this choice of parameterization increases the scatter on the graphs.

Figure 1a plots the partitioning data for As. Our As partition coefficients agree well with the limited previous experimental data [12]. Because of the greater incompatibility of As relative to Ni, recent studies have plotted elemental abundances against As to display iron meteorite fractionation trends [7]. An increased understanding of the effects of P and S on the solid metal/liquid metal partitioning of As will be useful to model and interpret these trends. The partitioning results for Ga are shown in Fig. 1b. Our new Ga data also agree nicely with previous work [2,3,11,13] and extend the range of S and P contents over which k(Ga) has been determined.

The sensitive determinations of partition coefficients of low-level doped runs allow a direct test of Henry's Law behavior. The measured concentration of As in the run products ranged from 0.2 wt% to 4ppm, Ga varied from 0.14 wt% to 0.9 ppm, and Ir ranged from 0.1 wt% to 6 ppb. For comparison, IIIAB iron meteorites contain 2-20 ppm As, 14-24 ppm Ga, and 10ppm-10ppb Ir [7]. These trace element concentration levels are significantly lower than previous experimental studies that used the electron microprobe. As shown in Fig. 1, our partitioning results are in good agreement with those previous studies.

There are fewer data points for Ir than As or Ga because Ir was below detection limits in some experiments. However, in Fig. 1c, many of our results were from experiments with Ir levels more than 3 orders of magnitude lower than would be possible for the electron microprobe. Despite the difference in Ir concentrations, our results agree well with the previous Ir studies [2,3,11,14,15]. These new k values complement existing studies, but importantly indicate that experimentally determined partition coefficients for siderophile trace elements are free of deviations from Henry's Law. This removes a large uncertainty involved in using experimental k values for modeling fractional crystallization in iron meteorites.

References: [1] Scott E. R. D. (1972) *GCA, 36*, 1205-1236. [2] Willis J. and Goldstein J. I. (1982) *LPSC, 13, I, JGR, 87*, A435-A445. [3] Jones J. H. and Drake M. J. (1983) *GCA, 47*, 1199-1209. [4] Haack H. and Scott E. R. D. (1993) *GCA, 57*, 3457-3472. [5] Ulff-Moller F. (1998) *MAPS, 33*, 207-220. [6] Chabot N. L. and Drake M. J. (1999) *MAPS, 34*, 235-246. [7] Wasson J. T. (1999*) GCA, 63*, 2875-2889. [8] Campbell A. J. and Humayun M. (1999) *Anal. Chem., 71*, 939-946. [9] Campbell A. J. et al. (2001) *GCA, 65*, 163-180. [10] Chabot N. L. and Drake M. J. (2000) *MAPS, 35*, 807-816. [11] Jones J. H. and Malvin D. J. (1990) *Met. Trans. B, 21B*, 697-706. [12] Jones J. H. *unpublished data*. [13] Hillgren V. J. (1993) *thesis*. [14] Malvin D. J. et al. (1986) *GCA, 50*, 1221-1231. [15] Fleet et al. (1999) *GCA, 63*, 2611-2622.
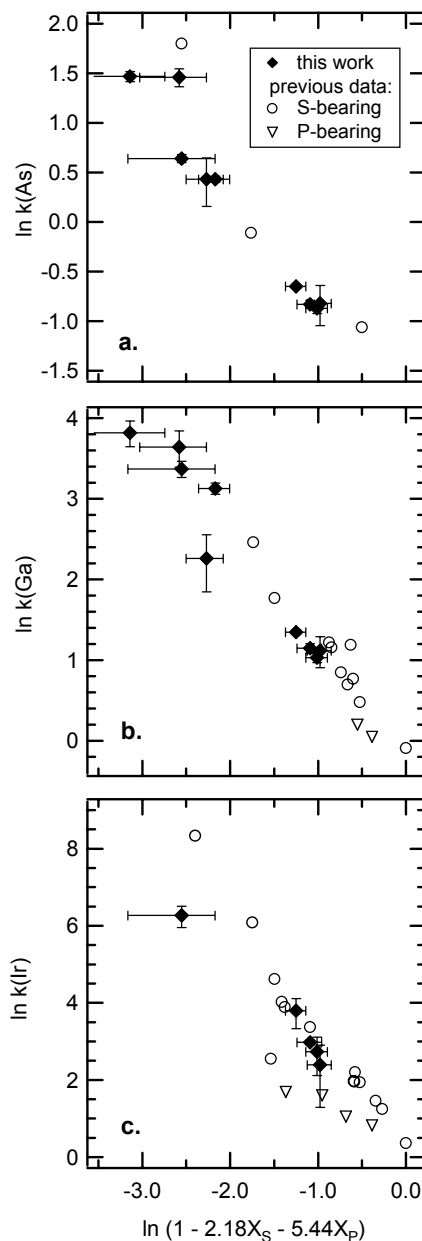
Fig. 1: The molar solid metal/liquid metal partition coefficients (k) for (**a.**) As, (**b.**) Ga, and (**c.**) Ir are plotted as a function of the molar concentration of S (X_S) and P (X_P) in the metallic liquid. Our data agree well with previous work, despite our lower levels of trace elements concentrations.