DIFFUSION-DRIVEN KINETIC ISOTOPE FRACTIONATION OF Fe AND Ni IN IRON METEORITES: A NEW DIMENSION TO THE ANALYSIS OF COOLING RATES. N. Dauphas[1], [1]Origins Laboratory, Department of the Geophysical Sciences and Enrico Fermi Institute, The University of Chicago, 5734 South Ellis Avenue, Chicago IL 60637, dauphas@uchicago.edu.

Introduction: Widmanstätten patterns in iron meteorites show concentration gradients indicating that growth of kamacite out of taenite occurred in a diffusion-limited regime. This feature can be used to estimate cooling rates [1,2]. Formation of Widmanstätten pattern is complex to model because many parameters are involved (*e.g.*, degree of undercooling when kamacite nucleation occurs, positions of the phase boundaries as a function of temperature and composition of the alloy, interdiffusion coefficients also as a function of temperature and composition of the alloy).

Isotopes can be fractionated during diffusive transport (light isotopes diffuse faster than heavy ones). Recently, Roskosz *et al.* [3] reported Fe isotopic fractionation during diffusion in Pt at 1,500 °C. They concluded that the diffusivities of adjacent isotopes of Fe differed by ~4 ‰/amu. If the same parameter applies to Fe and Ni diffusion in Fe-Ni alloy during growth of Widmanstätten pattern (~400-700 °C), then measurable kinetic isotope fractionation may be present in iron meteorites. Several research groups have reported Fe and Ni isotopic fractionation in adjacent taenite and kamacite [4-8], which can indeed be explained by diffusion-driven kinetic isotope fractionation [8,9].

Observations: The Toluca iron meteorite (IAB) is the most extensively studied sample for Fe and Ni isotopic fractionation [4-7] and cooling rate estimate [*e.g.*, 10]. For this reason, it is used here as a case example to compare modeling and observations. Measured Fe and Ni isotopic fractionations published to this day [4-7] are shown in Fig. 1. Taenite has similar or slightly heavy Fe isotopic composition relative to kamacite (0 to 0.1‰/amu). In contrast, taenite has significantly light Ni isotopic composition relative to kamacite (-0.4 ‰/amu). Both the directions and relative magnitudes of Fe and Ni isotopic fractionations are consistent with diffusion-driven kinetic isotope fractionation. Because taenite and kamacite have higher concentrations of Fe compared to Ni (by factors of 4 to 10), the diffusive signal should be diluted to a greater extent by normal (0 ‰/amu) background for Fe than for Ni. So a larger absolute fractionation is expected for Ni compared to Fe, which is observed. Growth of kamacite out of taenite is mainly limited by diffusion in taenite. In all studies reporting Fe and Ni isotopic analyses of iron meteorites, the sampling location was either not specified or of insufficient spatial resolution to be useful. Most likely, the samples analyzed came from the center regions of the phases. Because the center of taenite has low Ni concentration compared to the border, the net diffusive flux is from the border to the center and one would expect taenite analyses to show light Ni/heavy Fe isotopic compositions relative to kamacite. Again, this is exactly what is observed.

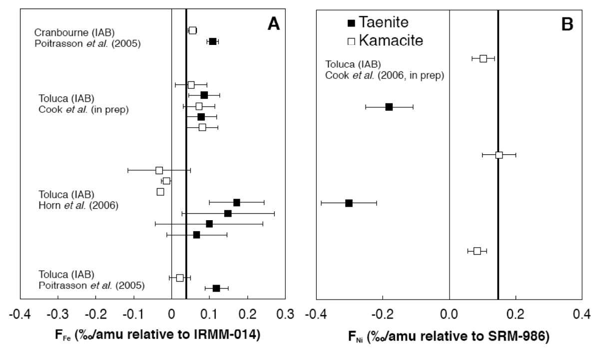

Fig. 1. Fe and Ni isotope measurements in taenite and kamacite of IAB iron meteorites. (A) Fe isotopic composition in ‰/amu relative to IRMM-014 [4,5,7] (B) Ni isotopic composition in ‰/amu relative to SRM-986 [6,7]. The vertical, heavy lines are the average Fe and Ni isotopic compositions of bulk iron meteorites.

Test of the model and results: Formation of Widmanstätten pattern was modeled using the phase diagram and interdiffusion coefficients of Hopfe and Goldstein [10]. The influence of P on the phase diagram was taken into account using the method of Moren and Goldstein [11]. Kamacite was assumed to nucleate at 700 °C [10]. The differential equations that govern the transport of Fe and Ni in taenite and kamacite and the movement of the interface were solved using a fixed finite-difference grid method with Lagrangian interpolation at the interface [12]. The algorithm was tested against Ni concentration profiles given by [10] and the analytical asymptotic self-similar solution of a problem that shares similarities with the problem at hand [13]. A new development of this study is the computation of the kinetic isotope fractionation associated with formation of Widmanstätten pattern. It does not directly follow from modeling concentration profiles because the isotopic composition at the interface in not known *a priori*. A β-factor $[D_2/D_1=(m_1/m_2)^{\beta}]$ similar to that obtained by Roskosz et al. [3] for Fe diffusion in solid Pt at 1,500 °C (0.25) was used here. Model results critically depend on this value, which is still uncertain.

Figure 2 shows model results for Ni using a cooling rate of 25 °C/Myr, a value derived by [10] using

Wood's method of matching taenite half-width and central Ni concentration [1]. A major feature is that at this cooling rate and at 450 °C, kamacite shows a significant difference in Ni isotopic composition between the interface and the center. The reason is that although the interdiffusion coefficients in kamacite are always higher than in taenite, the size over which atoms must diffuse becomes larger for kamacite as the crystal grows at the expense of taenite. The best match between measurements and modeling is obtained for a cooling rate of ~50 °C/Myr, which is within error bars similar to the value obtained by [10] using Ni profiles.

Conclusions and perspectives: The isotopic fractionation measured between taenite and kamacite in iron meteorites most likely results from diffusion-driven kinetic isotope fractionation. However, the key parameter that governs this process (β) is still unknown for the alloy composition and temperature investigated in this study. We also do not know whether there is any equilibrium fractionation at the interface between taenite and kamacite. With these caveats in mind, Fe and Ni isotopic fractionation adds a new dimension to the analysis of meteorite cooling rates. Very few extra-parameters need to be determined to extend algorithms used for computing Ni concentration profiles to include isotopes. Diffusion-driven kinetic isotope fractionation represents the most straightforward test of the assumptions and parameters that go into modeling formation of Widmanstätten pattern. It is worthwhile to note that isotopes and concentrations are not redundant. Concentration gradients can be present with no isotopic fractionation and *vice-versa*. Important targets for study are IVA iron meteorites, where Ni concentration profiles indicate that the meteorites cooled at different rates, which contradicts the idea that they formed in an isothermal asteroid core overlaid by a silicate mantle [*e.g.*, 11,14,15].

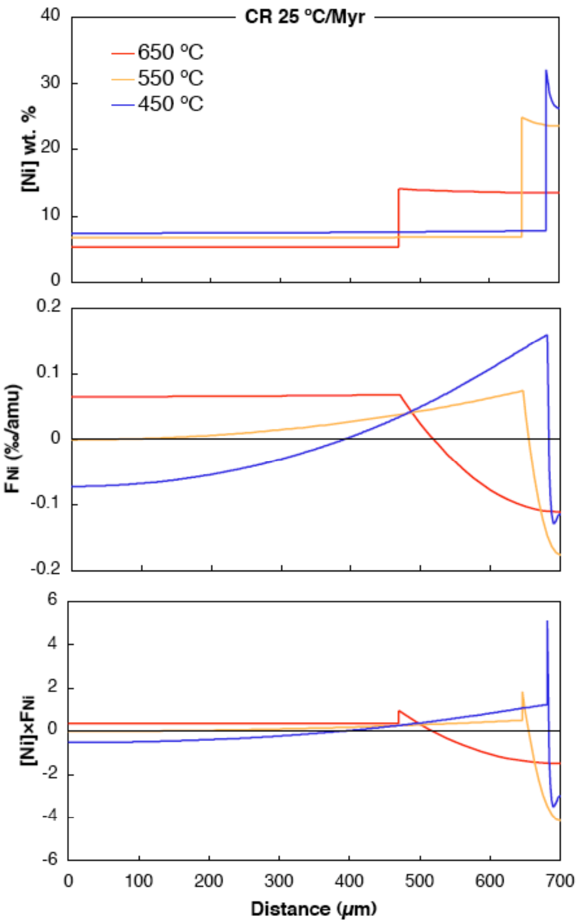

Fig. 2. Ni concentration and kinetic isotope fractionation (β=0.25). The three colored curves in each panel correspond to snapshots taken at different temperatures (650 ºC, red; 550 ºC, orange; 450 ºC, blue). Below 450 ºC, the system is almost frozen. The bottom panel shows the product of the Ni concentration and isotopic composition (the area below the curve must be equal to 0 to respect mass-balance).

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