NICKEL ISOTOPE SYSTEMATICS IN TROILITE FROM MAGMATIC AND NON-MAGMATIC IRON METEORITES. D. L. Cook[1,2,3], R. N. Clayton[1,2,4], M. Wadhwa[2,3,†], P. E. Janney[2,3,†], and A. M. Davis [1,2,4], [1]Department of the Geophysical Sciences, The University of Chicago, 5734 S. Ellis Ave., Chicago, IL 60637 (davecook@uchicago.edu), [2]Chicago Center for Cosmochemistry, 5640 S. Ellis Ave., Chicago, IL 60637, [3]Department of Geology, The Field Museum, 1400 S. Lake Shore Dr., Chicago, IL, 60605, [4]Enrico Fermi Institute, The University of Chicago, 5640 S. Ellis Ave., Chicago, IL, 60637, [†]Present address: Center for Meteorite Studies, School of Earth and Space Exploration, Arizona State University, P.O. Box 871404, Tempe, AZ 85287-1404.

Introduction: Investigations of the nickel isotope compositions of meteorites and their components can provide unique constraints on the formation time scales and the possible presence of nucleosynthetic anomalies in solids formed in the early solar system. Unambiguous evidence of excesses in radiogenic ^{60}Ni from the decay of ^{60}Fe ($t_{1/2}$ = 1.49 My) was first reported in bulk eucrite samples [1], which formed in differentiated parent bodies. More recently, analyses of various phases in unequilibrated enstatite [2] and ordinary [3-5] chondrites have revealed ^{60}Ni excesses in sulfides, oxides, and silicates. Additionally, deficits of ^{60}Ni have been reported in metal separates from several unequilibrated chondrites [6]. These deficits are consistent with metal formation from a chondritic reservoir when ^{60}Fe was extant. The deficits in metal [6] and the excesses in non-metal phases [4,5] in unequilibrated chondrites indicate that the solar system initial ^{60}Fe/^{56}Fe was at least $\approx 10^{-6}$.

Nucleosynthetic anomalies in Ni isotopes have been reported in some meteoritic components. In particular, anomalies in the neutron-rich isotopes of Ni were reported in several Allende CAIs [7]. Also, a recent study indicated the presence of possible nucleosynthetic anomalies in Ni isotopes in sulfides of several iron meteorites [8]; these anomalies are rather enigmatic since none were detected in metal from the same iron meteorites. To clarify the question of whether or not anomalies of nucleosynthetic origin are indeed present in sulfides of iron meteorites and if these phases record any evidence of live ^{60}Fe, we have analyzed the Ni isotopic compositions of troilite (FeS) from several iron meteorites

Samples and Methods: Samples were chosen to represent a wide variety of iron meteorite groups including magmatic and non-magmatic irons. Specifically, troilite from the following meteorites was analyzed: Canyon Diablo (IAB), Mundrabilla (IAB), Odessa (IAB), Toluca (IAB), Gressk (IIAB), Augustinovka (IIIAB), Bella Roca (IIIAB), Chupaderos (IIIAB), Grant (IIIAB), and Gibeon (IVA). We also analyzed three samples of terrestrial Fe-Ni sulfides to verify that there were no analytical artifacts resulting from our chemical separation and mass spectrometry protocol; these samples included pentlandite ($(Fe,Ni)_9S_8$), pyrrhotite ($Fe_{1-x}S$), and violarite ($FeNi_2S_4$). Troilite separates were available from the Field Museum collection for three of the iron meteorites studied. For the remaining samples, troilite nodules in iron meteorite slabs were sampled using stainless steel dental tools. Approximately 100 mg of troilite was obtained from each iron meteorite sample. All sulfides (terrestrial and meteoritic) were digested in Teflon beakers using reverse *aqua regia* (2:1 mix of conc. HNO_3 to conc. HCl). Nickel was separated using a combination of anion and cation exchange chromatography by modifying the protocol described by [6] for processing metallic samples. In addition to the natural samples, three aliquots of NIST SRM 986 Ni standard were processed through the Ni separation chemistry. The total procedural blank for Ni is ≈ 4.5 ng and is insignificant in comparison to the amount of Ni in the samples.

Ni isotope measurements were made at the Isotope Geochemistry Laboratory of the Field Museum using a Micromass IsoProbe multi-collector ICPMS. Samples were measured via the standard-sample bracket technique using the NIST SRM 986 as the Ni isotope standard. Samples were corrected for mass bias using an exponential law and ^{62}Ni/^{58}Ni ≡ 0.053388 [9]. In addition to measuring the five Ni isotopes, ^{57}Fe and ^{66}Zn were monitored and used to correct isobaric interferences on ^{58}Ni from ^{58}Fe and on ^{64}Ni from ^{64}Zn. A more detailed discussion of the analytical routine can be found in [6]. The Fe/Ni ratios of troilite samples were determined in sample solutions using a Varian ICPMS instrument equipped with a quadrupole mass analyzer at the Field Museum.

Precision and Accuracy: The data are reported in epsilon units, given as:

$$\varepsilon^i = [(R_{sample} - R_{standard})/R_{standard}] \times 10^4$$

where R is the mass bias corrected ^{i}Ni/^{58}Ni ratio (i = 60, 61, 64). Repeated analyses of an Aesar Ni solution bracketed with the NIST SRM 986 standard over a 27-month period were used to determine the external precisions for ε^{60}, ε^{61} and ε^{64}. The external precision (2SD) for ε^{60} based on a protocol consisting of 5 repeat measurements during a single analytical session is ± 0.15 ε, as previously re-

ported [6]. A protocol consisting of 10 repeat measurements during a single analytical session resulted in an improvement of 40% in the external precision for ε^{60} (± 0.09 ε). Similar improvements of 35% and 20% were also obtained for ε^{61} and ε^{64}, respectively. Since the latter protocol results in an improved ability to resolve potential anomalies in the Ni isotopes, we performed 10 repeat measurements for all troilite samples.

The measured ε^{60} (Fig. 1), ε^{61}, and ε^{64} values for the terrestrial sulfides and the processed aliquots of SRM 986 are all zero within uncertainty. Hence, these data demonstrate that our chemical separation and mass spectrometry procedures for sulfides are free from analytical artifacts.

Results and Discussion: Despite the fact that the troilites analyzed here had $^{56}Fe/^{58}Ni$ ratios up to ~8600, no resolvable excesses in ε^{60} from the decay of ^{60}Fe were measured in any of the samples. This is consistent with the results of previous studies of troilite from iron meteorites [8,10]. The lack of a resolvable excess in radiogenic ^{60}Ni and the corresponding Fe/Ni ratio in the troilite of each meteorite analyzed may be used to estimate the minimum time interval (ΔT_{min}) required between the beginning of the solar system, as defined by the formation of CAIs, and the formation time (*i.e.*, the time of cooling through the closure temperature of the Fe-Ni system) of that meteorite. The $^{56}Fe/^{58}Ni$ ratios of the troilites analyzed range from 54 (Odessa) to 8599 (Grant). Thus, if one assumes a solar system initial $^{60}Fe/^{56}Fe$ ratio of $\approx 10^{-6}$ [4-6], a ΔT_{min} of ≈ 4.5 to ≈ 17 My is estimated.

Whereas there are no resolvable excesses in ε^{60} in any of the sulfides analyzed here, there are significant deficits in at least 5 of the 10 troilites (Fig. 1). Although this is broadly consistent with the results of [8], there are some differences in detail. Specifically, while [8] reported the largest anomalies in ε^{60} (and ε^{61}; no ε^{64} data were reported by these authors) in Odessa troilites, we did not detect any anomalies in ε^{60}, ε^{61} or ε^{64} in our troilite sample from this same meteorite. In fact, we did not observe clearly resolvable anomalies in ε^{61} in any troilites, except for Mundrabilla troilite which has a slightly negative ε^{61}. Finally, we have detected anomalies in ε^{64} that appear to broadly correlate with the effects in ε^{60}. In particular, Mundrabilla, for which we observe the largest deficit in ε^{60}, also shows the largest deficit in ε^{64}. As was shown by [8], different troilite samples from the same meteorite (*e.g.*, Odessa and Toluca) showed distinct anomalies in ε^{60} and ε^{61}. As such, the above noted differences between the results of [8] and this study may be a record of real heterogeneities in the isotopic compositions of these troilites. The Ni isotopic compositions of the troilites measured by us are inconsistent with an excess or deficit of a pure s-process component preserved in these sulfides. At present, the origin of these anomalies in troilites from iron meteorites remains enigmatic.

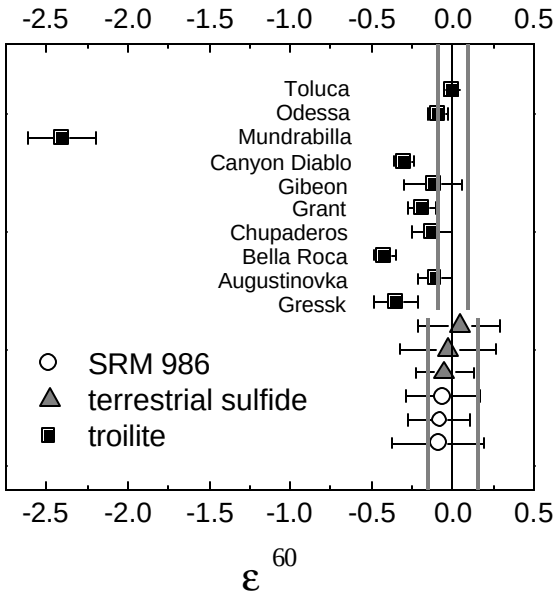

Fig. 1: ε^{60} values for aliquots of SRM 986, terrestrial sulfides, and meteoritic troilite processed using the Ni separation chemistry developed for sulfide samples. Each datum represents the mean of multiple repeat measurements (5 repeats for SRM 986 aliquots and terrestrial sulfides and 10 repeats for troilites). Error bars are $2\sigma_m$. The external precisions (2SD) for both protocols are shown by the two sets of solid gray lines.

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