

HIGH PRECISION NICKEL ISOTOPIC ANALYSES OF METEORITES. H. Tang¹ and N. Dauphas¹,
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Introduction: Among short-lived nuclides in meteorites, ⁶⁰Fe ($t_{1/2}=2.62$ My [1]) plays an important role as a chronometer and a heat source in early-formed planetary bodies [2]. Evidence for presence of ⁶⁰Fe has been found in eucrites in the form of excess ⁶⁰Ni [3,4]. Recently, the ⁶⁰Fe/⁵⁶Fe ratio in the early solar system was constrained more precisely by *in situ* analysis of high Fe/Ni phases in chondrules [5-8]. Initial ⁶⁰Fe/⁵⁶Fe ratios of $2.2\text{-}3.7 \times 10^{-7}$ in chondrules can be used to estimate an initial ratio of $5\text{-}10 \times 10^{-7}$ at the time of condensation of calcium-aluminum-rich inclusions CAIs (using a time interval of 1.5-2.0 My between formation of CAIs and chondrules [9,10]). High-precision measurements of ⁵⁸Fe, a neutron-rich isotope of iron produced together with ⁶⁰Fe in AGB-stars and supernovae, have shown that ⁶⁰Fe was homogeneously distributed at the scale of bulk planetary objects [11,12]. However, no direct evidence for the presence of ⁶⁰Fe has yet been detected in chondrites by MC-ICPMS or TIMS. Variations in ⁶⁰Ni have been detected but these correlate with variations of other stable Ni isotopes and may have a nucleosynthetic origin [13,14]. These anomalies are small and affect only certain meteorites. We developed a new procedure to purify nickel from chondrites and analyze its isotopic composition by MC-ICPMS in order to ascertain the presence of Ni isotopic anomalies reported by previous workers and to search for the evidence of the past presence of live ⁶⁰Fe in chondrites.

Separation of Ni for isotopic analysis: Iron meteorites were leached in 3 M HCl for several hours to remove surface contamination. They were then dissolved in a 1:2 mixture of HNO₃ and HCl. Chondrites, which contain chemically resistant phases, were digested by Parr bomb dissolution and flux fusion [15]. Nickel was separated from all samples using four steps of ion exchange chromatography. Attention was paid to devise a method that does not use dimethylglyoxime, a Ni-specific organic compound used in several studies. The first column is filled AG1W-X8 resin and separates Ni from Fe, Cu, Co and Ti [12]. The second column is filled with AG50W-X8 and separates Ni from lithophile elements like Na, Al, K and Li. The remaining matrix elements such as Ca, Mg and Mn can be removed in the third column filled with AG50W-X12 in a mixture of 80 % acetone + 20 % 10 M HCl [16]. This step is repeated several times. A fourth column is filled with AG1W-X8 resin and is used to eliminate Zn in HBr medium [17]. With the chemical purification procedures just mentioned, the intensities of major interferences such as Mg and Fe were reduced to the level of acid blanks. Isotopic analyses

were performed on a Neptune MC-ICPMS in high resolution to remove argon interferences from Fe and Ni isotopes. The level of ⁶⁴Zn interference on ⁶⁴Ni can be significant, therefore standards doped with Zn were analyzed with samples to optimize that correction. The measurements were replicated 20 times bracketed by SRM986. The uncertainties correspond to the 95 % confidence interval of the average.

Results and Discussion: Terrestrial standard (SRM986) passed through the columns has normal composition for ⁶⁰Ni, ⁶²Ni and ⁶⁴Ni within uncertainties (ϵ_{Ni} are 0.011 ± 0.048 , 0.006 ± 0.091 and 0.054 ± 0.167 , respectively). Two carbonaceous chondrites (Allende and Murchison) show significant $\epsilon^{60}\text{Ni}$ deficits as well as large $\epsilon^{62}\text{Ni}$ excesses and $\epsilon^{64}\text{Ni}$ excesses relative to enstatite chondrites (Khairpur and St Mark's), whose ϵ_{Ni} overlap with the terrestrial values within uncertainties. Two of the iron meteorite samples (Cape of Good Hope and Tlacotepec) have distinct Ni isotopic compositions from terrestrial Ni. Several iron meteorite samples that we analyzed before did not reveal any departure from terrestrial composition [11]. They also did not show any anomaly in the study presented by [13]. In Fig. 1, the data obtained here and [11] were plotted against the data in [13,14,18] from the same samples. Within uncertainties our results agree with [13,14,18]. The preliminary data presented here confirm the results presented by [13] that isotopic anomalies are present for nickel in bulk meteorites.

Small ⁶⁴Ni effects were observed in both carbonaceous chondrites and iron meteorites. These effects are at the limit of analytical precision and work is currently under progress to ascertain that these are real. If confirmed, these anomalies do not imply that ⁶⁰Fe was heterogeneously distributed in the early solar nebula [19]. Such anomalies could be carried by presolar grains unrelated to the star(s) that produced the ⁶⁰Fe found in meteorites. The measured heterogeneity in ⁶⁴Ni abundance in bulk meteorites constrains the heterogeneity of the initial ⁶⁰Fe/⁵⁶Fe ratio to less than $\pm 18\%$ dispersion around the average (using Eq. 3 of [11]), assuming that ⁶⁴Ni was coupled with ⁶⁰Fe and ⁵⁸Fe during the injection. This constraint is not as tight as that obtained using ⁵⁸Fe [11], which indicates that ⁶⁰Fe was homogeneously distributed at the $\pm 10\%$ level. Another advantage of ⁵⁸Fe compared to ⁶⁴Ni is that it cannot be chemically decoupled from ⁶⁰Fe. Therefore, ⁵⁸Fe will be analyzed in the same samples that we analyzed for this study.

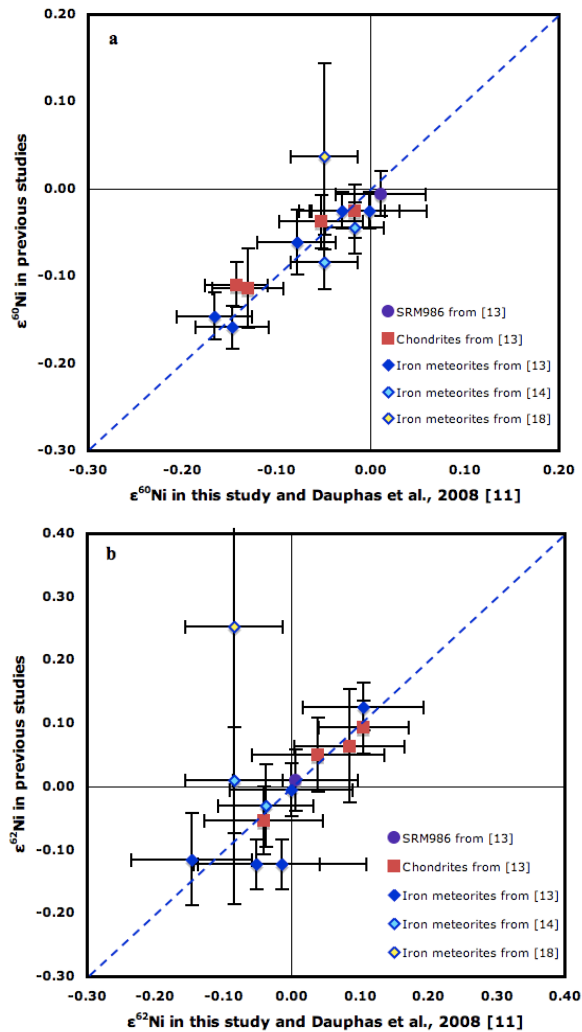


Fig.1. Ni isotope comparison between data from the Origins Laboratory (this study and [11]) and results from [13,14,18] for the same meteorites. The blue dash line is $y=x$.

Conclusion: High-precision data on nickel isotopes in meteorites agree with results from previous studies. Measurable Ni isotopic heterogeneity is present in bulk meteorite samples. The presence of heterogeneities in Ni stable isotopes complicates the search for live ^{60}Fe by TIMS or MC-ICPMS as one must find a way to distinguish variations in ^{60}Ni that arise from decay of ^{60}Fe from other effects. More meteorite groups as well as geostandards will be analyzed to reaffirm the accuracy of our data. The new method for nickel separation will be improved to also document mass-dependent fractionation in meteoritic [20] and terrestrial [21] materials.

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