CONSTRAINTS FROM ACHONDRITES ON THE INITIAL $^{60}Fe/^{56}Fe$ RATIO OF THE SOLAR SYSTEM.

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Introduction: Among short-lived nuclides inferred to have been present in meteorites, ^{60}Fe ($t_{1/2}$=2.62 Myr [1]) could play an important role as a chronometer and a heat source in early-formed planetary bodies [2]. The first evidence for the presence of ^{60}Fe in solar system material was found in eucrites in the form of excess ^{60}Ni, which defines an internal isochron [3,4]. However, the closure time for the Fe-Ni system was not well known, hindering a reliable estimate of the $^{60}Fe/^{56}Fe$ ratio at solar system formation. The initial ratio was constrained more precisely by *in situ* analyses of high Fe/Ni phases in chondrules [5-9]. Initial $^{60}Fe/^{56}Fe$ ratios of $2.2\text{-}3.7\times10^{-7}$ in chondrules correspond to an initial ratio of $(6.3\pm2.0)\times10^{-7}$ at the time of condensation of calcium-aluminum-rich inclusions (CAIs). However, this ratio is controversial owing to the possible presence of unresolved interferences on Ni isotopes [10]. Recently, the initial ratio was constrained using bulk and mineral analyses of angrites with high Fe/Ni ratios [11]. Quitté et al. obtained an initial $^{60}Fe/^{56}Fe$ ratio of $(3.12\pm0.78)\times10^{-9}$ at the time of angrite formation, which translates into a solar system initial ratio of $(7.47\pm3.05)\times10^{-9}$ at the time of CAI formation. They also argued that ^{60}Fe was heterogeneously distributed in the proto-planetary disk.

To address the question of the initial abundance of ^{60}Fe and of its distribution in the disk, we have measured the Ni isotopic compositions of bulk eucrites, thus avoiding uncertainties in the closure time of internal isochrons. In eucrites, high Fe/Ni ratios were produced by core-mantle segregation as well as magma ocean crystallization [12]. Core formation and mantle differentiation in the eucrite parent body has been suggested to have taken place within the first ~4 Myr of solar system formation based on Hf-W systematics [13, 14]. The $^{53}Mn\text{-}^{53}Cr$ system also constrains the time of magma ocean crystallization in the HED parent-body to 2.2±1.1 Myr after CAIs. Therefore, one would expect Fe/Ni fractionation in bulk eucrites to have occurred while ^{60}Fe was still present.

Although ureilites and aubrites have lower Fe/Ni ratios than eucrites and their chronology is not as well constrained, we have analyzed several samples to search for evidence of live ^{60}Fe in their parent-bodies.

Method for Ni Isotopic Analysis: Samples were cleaned with abrasive paper and rinsed in acetone to remove fusion crust. Because of the low Ni content of achondrites (~10ppm), ~0.5 g of material was digested. After digesting the samples in HF-HNO_3 and HCl-HNO_3 mixtures, nickel was separated from potential interferences and matrix elements using four steps of ion exchange chromatography. Attention was paid to devise a method that does not use dimethylglioxyme, a Ni-specific organic compound used in several studies. The first column is filled AG1W-X8 resin and separates Ni from Fe, Cu and Co [15]. The second column is based on a 2-mL TODGA cartridge and separates Ni from Ti, Hf and Zr [16]. Other matrix elements can be eliminated in the third column filled with AG50W-X12 in a mixture of acetone+HCl [17]. This step is repeated several times. Zinc is one of the most significant interferences on Ni, but it can be removed in a column filled with AG1W-X8 resin in HBr medium [18]. 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. A ~20 % aliquot was kept for the determination of Fe/Ni ratios. All concentration and isotopic measurements were performed on the Neptune MC-ICPMS of the Origins Lab in high resolution to remove argide interferences. Sample measurements were bracketed by SRM986. The measurements were replicated 6 - 17 times with the uncertainties corresponding to the 95 % confidence of the average.

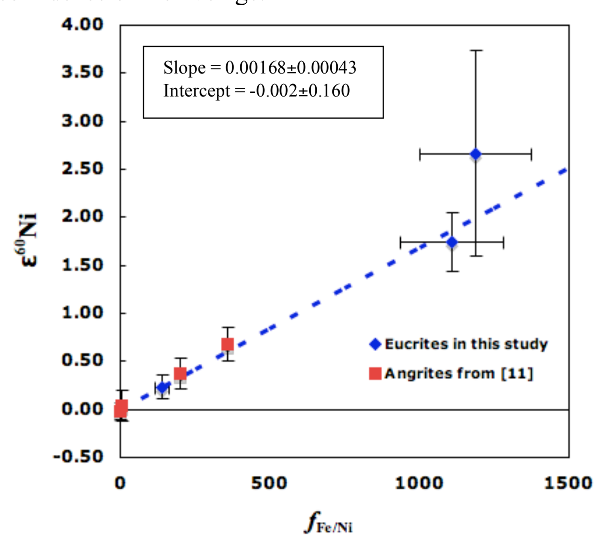

Figure 1. Fe-Ni isochron diagram for whole-rock basaltic eucrites and angrites. $\varepsilon^{60}Ni$ is the deviation of the $^{60}Ni/^{58}Ni$ ratio relative to the isotopic standard in part per 10,000 (mass fractionation was corrected by internal normalization using the $^{61}Ni/^{58}Ni$ ratio). $f_{Fe/Ni}$ is the Fe/Ni ratio normalized to the chondritic ratio.

Results: Terrestrial standards (AGV-02, BHVO-2 and GSP-02) that were passed through the columns have normal isotopic compositions for ^{60}Ni, ^{61}Ni and ^{62}Ni within uncertainties. Likewise, the ureilites (Kanna and EET 83309) and aubrite (Bishopville) investigated here, which have relatively low Fe/Ni ratios

(~100), have normal Ni isotopic compositions (εNi = 0) within analytical uncertainties. Three basaltic eucrites (Camel Donga, Juvinas and Ibitiria) show significant ε^{60}Ni excesses reaching ~+2 for a Fe/Ni ratio of ~20×10^3.

Discussion: *^{60}Fe/^{56}Fe initial ratio in the early solar nebula.* The Ni isotopic compositions of bulk eucrite samples correlate with their Fe/Ni ratios expressed as $f_{Fe/Ni}$ [$f_{Fe/Ni}$ = $(Fe/Ni)_{sample}/(Fe/Ni)_{CHUR}$-1] and define an isochron with a slope of 0.00168±0.00043 (Fig. 1). Based on the value of the slope, we estimate an initial ^{60}Fe/^{56}Fe ratio of $(2.74\pm0.70)\times10^{-9}$ at the time of core formation/magma ocean crystallization in the parent body of HEDs. According to ^{53}Mn-^{53}Cr whole-rock data for basaltic eucrites, mantle differentiation took place at 2.2±1.1 Myr after CAI formation [19]. We can thus calculate an initial ^{60}Fe/^{56}Fe ratio of $(4.90 \pm 2.68) \times 10^{-9}$ at the time of CAI formation.

The data of bulk angrites reported in [11] are also plotted in Fig. 1. The ^{60}Fe/^{56}Fe$_{initial}$ of the angrite parent body is estimated to be $(3.12 \pm 0.78) \times 10^{-9}$. Using the half-life of ^{60}Fe (2.62 Myr) and results from ^{53}Mn-^{53}Cr systematics of bulk angrites [20], we can estimate an initial ^{60}Fe/^{56}Fe ratio of $(7.47\pm3.05)\times10^{-9}$ at the beginning of the solar system. This is similar to the value that we derive from eucrites. There is a systematic difference however (by a factor of ~100) between the initial values derived from achondrites by MC-ICPMS and those derived from chondrites by SIMS. Further work will tell whether such discrepancies reflect heterogeneous distribution of ^{60}Fe, although this is highly unlikely in light of the homogeneity of ^{58}Fe abundences in solar materials [21].

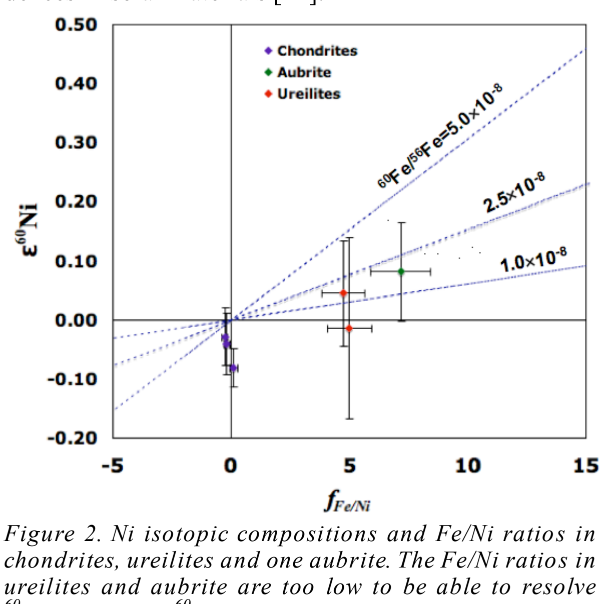

Figure 2. Ni isotopic compositions and Fe/Ni ratios in chondrites, ureilites and one aubrite. The Fe/Ni ratios in ureilites and aubrite are too low to be able to resolve ^{60}Ni excess from ^{60}Fe decay.

Ni isotopic composition in ureilites and aubrites. Results of bulk ureilites agree with previous work [11, 22]. Quitté et al. [11] argued that the lack of excess ^{60}Ni indicated that ^{60}Fe was heterogeneously distributed. However, based on our measurements, the precision of Ni isotopic analyses is not good enough to resolve any excess ^{60}Ni in bulk achondrites with Fe/Ni ratio lower than ~280 ($f_{Fe/Ni}$ < 15 in Fig. 2).

Conclusion: Evidence for live ^{60}Fe was found in bulk eucrites, from which we estimate the initial ^{60}Fe/^{56}Fe ratio at the time of CAI formation to be $(4.90 \pm 2.68) \times 10^{-9}$. This is identical within error to the initial ratio inferred from bulk angrite data, although we are currently analyzing more eucrites and angrites to further constrain the initial abundance of ^{60}Fe in the solar system.

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