

^{60}Fe - ^{60}Ni SYSTEMATICS OF CHAINPUR CHONDRULES AND THE PLUTONIC ANGRITES NORTHWEST AFRICA 4590 AND 4801. L. J. Spivak-Birndorf¹, M. Wadhwa¹, and P. E. Janney¹. ¹School of Earth and Space Exploration, Arizona State University, Tempe, AZ 85287 (Lev.Spivak-Birndorf@asu.edu)

Introduction: At present, the initial abundance and distribution of ^{60}Fe in the Solar System is not well constrained. Recent studies of the ^{60}Fe - ^{60}Ni systematics in whole-rock fractions and mineral separates from various achondrites, using multicollector inductively coupled plasma spectrometry (MC-ICPMS) suggest an initial Solar System $^{60}\text{Fe}/^{56}\text{Fe}$ ratio of $\sim 1 \times 10^{-8}$ [1-3]. In contrast, investigations of the ^{60}Fe - ^{60}Ni systematics in chondritic components using *in situ* measurements by secondary ion mass spectrometry (SIMS) suggest the initial $^{60}\text{Fe}/^{56}\text{Fe}$ ratio was $\sim 5\text{-}10 \times 10^{-7}$ [4, 5]. Furthermore, there is debate regarding whether or not the ^{60}Fe was distributed homogeneously in the early Solar System [2, 6]

Here we report on an investigation of the ^{60}Fe - ^{60}Ni systematics of bulk chondrules from the Chainpur unequilibrated ordinary chondrite (UOC). The goals of the study are to better constrain the initial Solar System abundance of ^{60}Fe and to obtain ^{60}Fe - ^{60}Ni systematics by MC-ICPMS in the same objects (i.e., chondrules in UOCs) that have previously been investigated by SIMS to allow evaluation of the possible reasons for the discrepant results obtained by these two techniques [4, 5]. Furthermore, we recently began an investigation of the ^{60}Fe - ^{60}Ni systematics of angrites to constrain the abundance and distribution of ^{60}Fe in the early Solar System [1, 7]. Here we report additional ^{60}Fe - ^{60}Ni data for two of the plutonic angrites Northwest Africa (NWA) 4590 and 4801.

Samples and Methods: Four chondrules weighing between ~ 2 and ~ 9 mg were separated from the Chainpur UOC. The chondrules were gently abraded with alumina polishing paper to remove adhering matrix and then ultrasonicated in ultra-pure water for ~ 10 min. The chondrules were dried, re-weighed and then digested in a 3:1 HF-HNO₃ mixture.

Bulk interior fragments of the plutonic angrites NWA 4801 and NWA 4590 were gently crushed with a boron carbide mortar and pestle. From this crushed material, one whole-rock fraction each of NWA 4590 (~ 200 mg) and NWA 4801 (~ 125 mg) was ultrasonicated in ultra-pure water for ~ 10 min, and an additional whole-rock fraction of NWA 4590 (~ 200 mg) was ultrasonicated in 0.5 M HCl for the same amount of time. After decanting the wash solutions, the samples were dried, re-weighed and digested. The bulk samples of NWA 4590 and NWA 4801 analyzed previously in our laboratory for ^{60}Fe - ^{60}Ni systematics [1] were leached in 0.05 M HCl without crushing prior to dissolution.

A $\sim 5\%$ aliquot of each sample solution was reserved for Fe/Ni ratio measurements, and the rest was chemically processed for Ni separation. The Fe/Ni ratios in chemically unprocessed solutions and the Ni isotopic compositions of the purified Ni from each of the samples were measured in the Isotope Cosmochemistry and Geochronology Laboratory (ICGL) at Arizona State University. Further details of the procedures for the measurement of Ni isotopes and Fe/Ni ratios have been described previously [1, 7].

Results: Chainpur chondrules. The Chainpur chondrules measured here have $^{56}\text{Fe}/^{58}\text{Ni}$ ratios ranging from ~ 140 to 575. All of the chondrules have Ni isotope compositions that are the same as the terrestrial composition within analytical uncertainties (i.e., $\epsilon^{60}\text{Ni}^* = 0 \pm 0.16\epsilon$ (2σ)). If the Chainpur chondrules are assumed to have formed contemporaneously, an upper limit on the $^{60}\text{Fe}/^{56}\text{Fe}$ ratio of $< 3.4 \times 10^{-8}$ can be estimated at this time.

Plutonic angrites. The two whole-rock samples of NWA 4801 (one washed in 0.05 M HCl and the other in ultrapure water) both have $^{56}\text{Fe}/^{58}\text{Ni}$ ratios of ~ 1000 . The two acid-washed whole-rock samples of NWA 4590 have $^{56}\text{Fe}/^{58}\text{Ni}$ ratios of ~ 9000 , while the whole-rock sample of NWA 4590 washed only in ultrapure water has a $^{56}\text{Fe}/^{58}\text{Ni}$ ratio of ~ 4000 . All of the plutonic angrite samples have Ni isotope compositions that are indistinguishable, within the errors, from the terrestrial composition (Fig. 1). If NWA 4590 and NWA 4801 are assumed to have crystallized at the same time [8, 9], we can estimate an upper limit on the $^{60}\text{Fe}/^{56}\text{Fe}$ ratio of $< 9 \times 10^{-10}$ when Ni isotopes in these angrites were last equilibrated.

Discussion: A recent study of the ^{53}Mn - ^{53}Cr systematics of bulk Chainpur chondrules estimated a $^{53}\text{Mn}/^{55}\text{Mn}$ ratio of $(5.1 \pm 1.6) \times 10^{-6}$ at the time of their formation [10]. This corresponds to a Mn-Cr age for Chainpur chondrules of 4565.8 ± 1.7 Ma, anchored to the ^{53}Mn - ^{53}Cr systematics [11] and U isotope corrected Pb-Pb age for D'Orbigny [12, 13]. Based on the lower limit of this Mn-Cr age for Chainpur chondrules, the maximum time difference between their formation and that of CAIs is ~ 4.3 Ma (relative to the oldest Pb-Pb age determined for a CAI [14]). The upper limit on the $^{60}\text{Fe}/^{56}\text{Fe}$ ratio for Chainpur chondrules determined here and this age difference can be used to estimate a maximum initial Solar System $^{60}\text{Fe}/^{56}\text{Fe} < \sim 1 \times 10^{-7}$. This value is ~ 5 to 10 times lower than estimates of the initial Solar System $^{60}\text{Fe}/^{56}\text{Fe}$ ratio from *in situ* SIMS studies of chondrites and their components [4, 5], but

is consistent with the lower values estimated based on analyses of achondritic meteorites by MC-ICPMS.

The lack of resolvable $^{60}\text{Ni}^*$ excesses in Chainpur chondrules measured here is also consistent with the limited data previously reported for bulk chondrules from UOCs [2, 15]. However, the chondrules analyzed here have the largest spread in Fe/Ni ratios measured so far (with $^{56}\text{Fe}/^{58}\text{Ni}$ ratios up to 575), and currently provide the strictest upper limit on the initial $^{60}\text{Fe}/^{56}\text{Fe}$ ratio estimated from bulk chondrules. The reason for the apparent discrepancy with the *in situ* SIMS chondrule data is not clear at this time and requires further MC-ICPMS measurements of mineral separates from individual UOC chondrules to obtain Fe-Ni internal isochrons.

A recent investigation of the ^{60}Fe - ^{60}Ni systematics of angrites [3] suggested that bulk samples of both quenched and plutonic angrites define an isochron corresponding to the time of widespread Fe/Ni fractionation in the mantle of the angrite parent body. However, that study included only two quenched angrites (D'Orbigny and Sahara 99555) and one plutonic angrite (NWA 2999, that has nearly chondritic ^{60}Fe - ^{60}Ni systematics) [3]. Therefore, it is not clear if these data for bulk angrite samples define a true whole-rock ^{60}Fe - ^{60}Ni isochron dating the timing of mantle Fe/Ni fractionation or correspond to the formation time of the parent magmas of the quenched angrites from a chondritic reservoir. More recently, additional Fe-Ni data were presented for whole-rock samples of the plutonic angrites [2], some of which showed resolvable $^{60}\text{Ni}^*$ excesses. In contrast, none of the whole-rock fractions (water or acid-washed) of the NWA 4590 and NWA 4801 plutonic angrites studied here show any resolvable radiogenic Ni excesses. This discrepancy is not likely to be due to terrestrial Ni contamination because all of the plutonic angrite samples measured in this study have Ni isotope compositions that are the same as the terrestrial composition regardless of the wash solvent (ultra-pure water, 0.05 M HCl or 0.5 M HCl) and amount of Ni (ranging from <0.5% to ~6% of the total) removed by washing. It is possible that the plutonic angrites have heterogeneous Ni isotope compositions (perhaps resulting from differing degrees of subsolidus isotopic re-equilibration in different portions of these samples), but this possibility remains to be investigated further.

The lack of resolvable $^{60}\text{Ni}^*$ excesses in whole-rock fractions of the plutonic angrites NWA 4801 and NWA 4590 reported here (with $^{56}\text{Fe}/^{58}\text{Ni}$ ratios from ~1000 to 9000) suggests that the parent magmas of these meteorites formed after the decay of ^{60}Fe . Therefore, the ^{60}Fe - ^{60}Ni systematics of bulk angrites do not reflect widespread contemporaneous differentiation of

their sources as proposed by [3], and instead correspond to the later formation ages of the plutonic angrites relative to the quenched angrites (consistent with the Pb-Pb ages for these samples [12]). The upper limit on the initial $^{60}\text{Fe}/^{56}\text{Fe}$ ratio based on the two whole-rock fractions of NWA 4801 and three whole-rock fractions of NWA 4590 corresponds to an age of <4557.4 Ma relative to D'Orbigny, which is in agreement with the absolute Pb-Pb ages reported for these meteorites [8, 9].

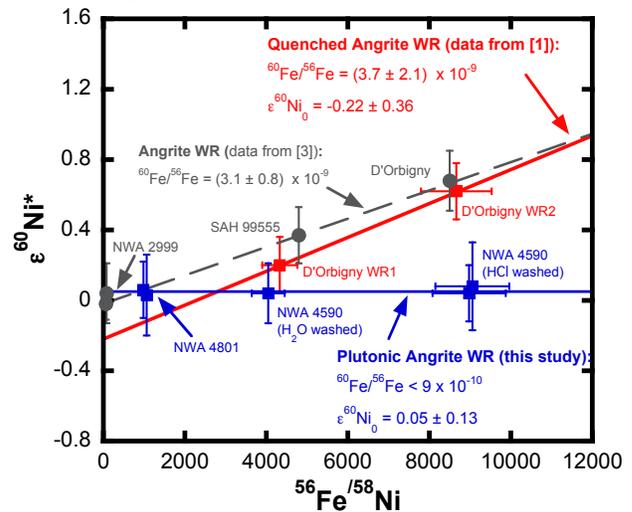


Fig. 1. Angrite whole-rock ^{60}Fe - ^{60}Ni systematics. Blue squares are whole-rock fractions of the plutonic angrites NWA 4590 and NWA 4801 (this study); Red squares are whole-rock fractions of the D'Orbigny quenched angrite [1]; Gray circles are whole-rock data from [3] for comparison.

References: [1] Spivak-Birndorf L. J. et al. (2011) *Workshop on Formation of the First Solids in the Solar System*, Abstract #9130. [2] Tang H. and Dauphas N. (2011) *Workshop on Formation of the First Solids in the Solar System*, Abstract #9146. [3] Quitté G. et al. (2010) *Astrophys. J.*, 720, 1215-1224. [4] Mostefaoui S. et al. (2005) *Astrophys. J.*, 625, 271-277. [5] Tachibana S. et al. (2006) *Astrophys. J.*, 639, L87-L90. [6] Quitté G. and Albarède F. (2011) *Workshop on Formation of the First Solids in the Solar System*, Abstract #9123. [7] Spivak-Birndorf L. J. et al. (2011) *LPS XLII*, abstract #2281. [8] Amelin Y. and Irving A. (2007) *Workshop on Chronology of Meteorites & the Early Solar System*, Abstract #4061. [9] Amelin Y. et al. (2011) *LPS XLII*, abstract #1682. [10] Yin Q. -Z. et al. (2007) *Astrophys. J.*, 662, L43-L46. [11] Glavin D. P. et al. (2004) *Meteorit. & Planet. Sci.*, 39, 693-700. [12] Amelin Y. (2008) *Geochim. Cosmochim. Acta*, 72, 221-232. [13] Brennecka G. A. and Wadhwa M. (2011) *Mineral. Mag.*, 75, 579. [14] Bouvier A. and Wadhwa M. (2010) *Nature Geosci.*, 3, 637-641. [15] Chen J H. et al. (2009) *Geochim. Cosmochim. Acta*, 73, 1461-1471.