NICKEL ISOTOPE ANOMALIES IN METEORITES AND THE ^{60}Fe-^{60}Ni CLOCK. M. Bizzarro[1], J.-L. Birck[2], J. Chen[3], G. Huss[4], G. Lugmair[5], S. Mostefaoui[6], D. Papanastassiou[3], A. Shukolyukov[5], G. Quitté[7], S. Tachibana[8], M. Wadhwa[9].

[1]Geological Institute, University of Copenhagen, Copenahgen, Denmark. [2]Laboratoire de Géochimie et Cosmochimie, IPG, Paris, France. [3]JPL, Pasadena, CA 91109-8099, USA. [4]HIGP, University of Hawai'i at Manoa. [5]Scripps Institution of Oceanography, University of California, San Diego, La Jolla, CA 92093–0212, USA. [6]LEME, Muséum National d'Histoire Naturelle, Paris, France. [7]Laboratoire des Sciences de la Terre, ENS, Lyon, France. [8]Department of Earth and Planetary Science, University of Tokyo. [9]Center for Meteorite Studies, School of Earth and Space Exploration, Arizona State University, Tempe, AZ 85287, USA.

Introduction: With a half-life of ~1.5 Myr, the ^{60}Fe-^{60}Ni decay scheme is ideally suited for dating meteorites and planetary processes that occurred in the first 10 Myr of the solar system's evolution. Moreover, ^{60}Fe is efficiently produced only by stellar nucleosynthesis such that constraining the timing of its first appearance, intitial abudance and distribution can constrain models of solar system formation. Ni possesses two neutron-rich isotopes, ^{62}Ni and ^{64}Ni, believed to be produced through nuclear statistical equilibrium processes occurring in neutron-rich supernova ejecta [1]. Both excesses and deficits have been documented for iron-group neutron-rich isotopes (^{48}Ca, ^{50}Ti, ^{54}Cr, ^{62}Ni and ^{64}Ni) in normal and FUN Ca,Al-rich inclusions (CAIs) as well as in primitive and differentiated meteorites [2-4].

Initial solar system abundance of ^{60}Fe: Hints of ^{60}Fe in the solar system first came from excesses of ^{60}Ni (^{60}Ni*) in CAIs with an inferred initial solar system ^{60}Fe/^{56}Fe [(^{60}Fe/^{56}Fe)$_0$] value of ~1.5 × 10^{-6} [5]. However, the evidence that ^{60}Fe was present at time of CAI formation remains ambiguous, given the lack of correlation of ^{60}Ni* values with Fe/Ni ratios and the presence of anomalies in other Ni isotopes. The first clear evidence for live ^{60}Fe in the solar system was discovered in basaltic meteorites believed to have formed at the surface of the eucrite parent body (EPB; [6-7]). The inferred (^{60}Fe/^{56}Fe)$_0$ from these meteorites is low enough to be consistent with an initial solar system ^{60}Fe abundance resulting from long-term galactic nucleosynthesis [8]. However, this interpretation is hampered by the extended and complex thermal history of meteorites originating from the EPB [9]. Initial attempts to search for traces of ^{60}Fe in more pristine objects such as primitive chondrite meteorites using *in situ* methods (i.e., secondary ionization mass spectrometry; SIMS) were unsuccessful [10]. With continuous effort and improvement in analytical methods, clear evidence for the former presence of ^{60}Fe in chondritic components was reported in troilite and magnetite [11,12]. These minerals yielded inferred (^{60}Fe/^{56}Fe)$_0$ ratios ranging from (1-1.8) × 10^{-7} for sulfides from the Bishunpur and Krymka (LL3.1) chondrites [11] to ~10^{-6} for sulfides from Semarkona (LL3.0) [12]. Given that the ^{60}Fe-^{60}Ni systematics in sulfides can be easily disturbed by mild thermal metamorphism or aqueous alteration, recent attempts to determine the initial solar system abundance of ^{60}Fe have focused on silicate materials [13]. Ferromagnesian pyroxene-rich chondrules from Bishunpur and Semarkona yielded inferred (^{60}Fe/^{56}Fe)$_0$ ranging from (2.2 ± 1.0) × 10^{-7} to (3.7 ± 1.9) × 10^{-7}. By applying the time difference of 1.5-2.0 Myr between formation of these chondrules and CAIs inferred from ^{26}Al-^{26}Mg systematics, a solar sytem (^{60}Fe/^{56}Fe)$_0$ of (5-10) × 10^{-7} is derived. This new estimate is inconsistent with the predicted steady state abundance of ^{60}Fe in the interstellar medium [8], and requires that a nearby stellar source interacted with the nascent solar system. Current models propose that either a supernova or a thermal pulsating asymptotic giant branch (AGB) star of intermediate mass produced and delivered ^{60}Fe to the solar system [13].

High-precision Ni isotopes measurements: With the advent of multiple collection inductively coupled plasma source mass spectrometry (MC-ICPMS) as well as second-generation thermal ionization mass spectrometry (TIMS), a number of laboratories have developed analytical protocols for high-precision measurements of Ni isotopes [14-18]. The precision and accuracy of these measurements, however, can be limited by the small size of three of the Ni isotopes, ^{61}Ni, ^{62}Ni and ^{64}Ni, with relative abundances of 1.1%, 3.6% and 0.9%, respectively. This is further exacerbated by the extensive number of potential isobaric interferences on the Ni mass array (~400) that may compromise the data. Currently, two approaches are taken to correct for instrumental mass bias during acquisition of the Ni isotope data. Some groups use the ^{62}Ni/^{58}Ni normalizing pair [14-16], while others use the ^{61}Ni/^{58}Ni ratio [17, 18]. Using the ^{62}Ni/^{58}Ni can provide better precision and accuracy given the higher natural abundance of ^{62}Ni, but the possible presence of nucleosynthetic anomalies at this mass may introduce biases in the ^{60}Ni/^{58}Ni data. Although the ^{61}Ni/^{58}Ni ratio may be potentially free of nucleosynthetic effects

[5], the intregrity of the data obtained using this normalization procedure is more easily compromised by the presence of any minor isobaric interferences, given the small natural abundance of ^{61}Ni.

Fossil ^{60}Ni* in CAIs? Re-investigations of Ni isotopes in CAIs [19] have confirmed the presence of correlated anomalies in ^{60}Ni and ^{62}Ni reported by [5]. Based on these data, a minimum solar system $(^{60}Fe/^{56}Fe)_0$ of 4.8×10^{-6} have been recently proposed [19], which is significantly higher than estimates derived from Bishunpur and Semarkona chondrules [13]. Two lines of evidence, however, suggest that the ^{60}Ni excesses present in CAIs may not record Fe/Ni fractionation at the time of CAI formation: (a) CAIs have subchondritic Fe/Ni ratios such that if these formed during the lifespan of ^{60}Fe, *deficits* in ^{60}Ni* should be observed, not excesses and (b) the ^{60}Ni* anomalies are not correlated with the Fe/Ni ratios. A tentative explanation is that ^{60}Ni excesses do not represent *in situ* decay of ^{60}Fe, but fossil ^{60}Ni* admixed to the CAI-forming reservoir. One of the required astrophysical environments for the nucleosynthesis of neutron-rich iron-group isotopes such as ^{48}Ca is the rare type Ia supernova having masses close to the Chandrasekar limiting mass [20]. These 'old' supernova sources produce ^{60}Fe, but no significant amount of ^{26}Al [21]. Therefore, admixing of this presolar component to the CAI-forming reservoir can generate correlated effects between ^{60}Ni* and neutron-rich iron group isotopes, without addition of ^{26}Al. In light of these caveats, we suggest that the CAI data may not accurately constrain the initial solar system abundance of ^{60}Fe.

Iron meteorites and pallasites: Recently published Ni isotope data for Fe-Ni metals and sulfides from iron meteorites and pallasites have yielded somewhat conflicting results. Using the ^{62}Ni/^{58}Ni for internal normalization, Quitté et al. [14] reported no ^{60}Ni anomalies at the ± 30 ppm level in Fe-Ni metals from iron meteorites, but significant correlated effects in ^{61}Ni and ^{60}Ni (up to ~1400 ppm) in co-existing sulfides, interpreted as nucleosynthetic anomalies. Cook et al. [15] and Chen et al. [16] reported no ^{60}Ni anomalies at the ± 20 ppm level for Fe-Ni metals. However, in contrast to [14], a normal Ni isotope composition was recently reported for sulfides from various irons by [16]. Using the ^{61}Ni/^{58}Ni for internal normalization, Bizzarro et al. [18] reported small deficits in ^{60}Ni of ~25 ppm with correlated effects on ^{62}Ni for Fe-Ni metals from iron meteorites and pallasites. Re-normalizing the data of [18] to ^{62}Ni/^{58}Ni yields a terrestrial ^{60}Ni composition to ±10 ppm and small resolvable ^{61}Ni excesses of ~30 ppm for these samples. We note that a preliminary report [17], using the same normalization procedure as [18] but with somewhat higher precision, also argues for the presence of widespread ^{62}Ni anomalies. Although there are contrasting systematics between the results obtained from several groups, the iron meteorite and pallasite data do not provide clear evidence for live ^{60}Fe at the time of Fe/Ni fractionation on their repective parent bodies.

Chondritic meteorites: Using the ^{62}Ni/^{58}Ni for internal normalization, Cook et al. [15] reported deficits in ^{60}Ni of ~25 ppm in metals separated from unequilibrated ordinary (OC) and carbonaceous chondrites (CC), including the well-dated Gujba chondrite [22]. If these reflect Fe/Ni fractionation from a chondritic reservoir during the lifespan of ^{60}Fe, the inferred $(^{60}Fe/^{56}Fe)_0$ at the time of Fe/Ni fractionation ranges from $(1.0\pm0.6) \times 10^{-6}$ to $(3.5\pm3.0) \times 10^{-6}$, which could be consistent with previous estimates based on Fe-Ni systematics in chondritic components [12,13]. Using the ^{61}Ni/^{58}Ni for internal normalization, Bizzarro et al. [18] reported a normal ^{60}Ni to ±15 ppm for bulk CC, OC and enstatite chondrites (EC), and small excesses in ^{62}Ni for CC, small deficits for OC, and normal ^{62}Ni for EC. These later results are in agreement with [17].

The SAH99555 angrite: With a well constrainted Pb-Pb age of 4564.55 ± 0.16 Myr [23], the SAH99555 angrite apparently provides a unique opportunity to anchor the ^{60}Fe-^{60}Ni system. Assuming a conservative solar system $(^{60}Fe/^{56}Fe)_0$ of 5×10^{-7}, crystallization of the SAH99555 angrite <5 Myr of CAIs would results in large excesses of at least 6 ε-units in the bulk samples. However, two different studies did not find the expected ^{60}Ni excesses, but reported either a terrestrial Ni isotope composition [24] or small ^{60}Ni and ^{62}Ni deficits [18]. Thus, these results are not consistent with estimates from Bishunpur and Semarkona chondrules.

Conclusion: Taken at face value, the currently available data for various meteorites does not provide compelling evidence for widespread distribution of ^{60}Fe in the early solar system and, therefore, does not support the chronological significance of the ^{60}Fe-^{60}Ni system. This could reflect either heteronegous distribution of ^{60}Fe in the solar system's parental molecular cloud or, alternatively, a late addition of ^{60}Fe to the protoplanetary disk.

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