**A REASSESSMENT OF THE $^{60}Fe$-$^{60}Ni$ ISOTOPIC SYSTEM AS A UBIQUITOUS CHRONOMETER FOR THE EARLY SOLAR SYSTEM.**

G. Quitté[1,4], A. Markowski[1], A.N. Halliday[2], M. Meier[1], C. Latkoczy[3], D. Günther[3], Ph. Telouk[4], J. Blichert-Toft[4] and F. Albarède[4], [1]IGMR, ETH Zurich, Switzerland. Email: Ghylaine.Quitte@ens-lyon.fr, [2]Dep. Earth Sciences, Oxford University, U.K., [3]Lab. Inorganic Chemistry, ETH Zurich, Switzerland, [4]LST, ENS Lyon, France.

**Introduction:** The short-lived $^{60}Fe$-$^{60}Ni$ chronometer ($t_{1/2}$ = 1.49 Myrs) can theoretically provide strong constraints on the exact chronology of the earliest events of the solar system. However, the expected anomalies are small in most cases because the Fe/Ni ratio is not strongly fractionated. This difficulty has been somewhat offset by the recent application of MC-ICPMS to Ni isotope analysis, which has helped improve the precision on this type of measurement. Another difficulty related to the Fe-Ni system is its mobility during metamorphism.

Over the past few years, we have analyzed Ni isotopes in different objects: Ca-Al-rich inclusions, iron meteorites, eucrites, and more recently angrites, to try to assess the early chronology of the solar system as inferred from the $^{60}Fe$-$^{60}Ni$ system and to establish whether this extinct radioactivity can be used as a chronometer for all types of objects.

**Mass spectrometry measurements:** Nickel was first extracted from the matrix elements following the procedure described by [1]. The isotopic composition was then determined using a large-geometry, high-resolution Nu 1700 MC-ICPMS at ETH Zurich. Angrite fractions were also measured on the Nu Plasma HR at ENS Lyon and the two data sets are in excellent agreement. Before analysis, the sample residue was taken up in either 0.1N HCl or 0.05N $HNO_3$. We tested that running samples in HCl versus $HNO_3$ medium makes no difference as far as signal stability, background level, washing time, and external reproducibility of the standard are concerned. The samples were thus run in HCl. Mass scans at a high mass resolution (8000-10000) revealed no interferences on the Ni masses except for small $ArNe^+$ and $ArO^+$ peaks in some cases. The mass fractionation was corrected by normalizing the $^{62}Ni/^{58}Ni$ ratio to 0.05338858 using an exponential law. Alternatively, data have also been normalized relative to $^{61}Ni/^{58}Ni$.

**Data processing:** There are different options for processing the data. First, the isotopic composition can be given as the average of all repeat measurements for a given sample and the uncertainty then corresponds to twice the standard deviation (2 SD) of all replicates taken together. Doing so, the external reproducibility achieved for an angrite typically is 0.3 ε and 0.8 ε for $^{60}Ni/^{58}Ni$ and $^{61}Ni/^{58}Ni$, respectively, normalized to $^{62}Ni/^{58}Ni$. Second, 3 measurements for each meteorite can be averaged and the mean value calculated. In this case, the external reproducibility becomes 0.2 ε and 0.6 ε for $^{60}Ni/^{58}Ni$ and $^{61}Ni/^{58}Ni$, respectively. As each sample has been measured more than 10 times, the uncertainty can also be reported as the standard error (2 SE), which can be as low as 0.08ε and 0.25 ε, respectively, for $^{60}Ni/^{58}Ni$ and $^{61}Ni/^{58}Ni$. The isotopic data presented below are average values of all repeat measurements and the uncertainty corresponds to twice the standard deviation (2 SD) of all replicates. Other groups prefer a different approach. The exact value of the reported uncertainties is extremely important for Ni isotopic measurements because the expected isotopic anomalies are so small for most samples. Depending on the reported error bars, the interpretation can vary significantly. For example, iron meteorites have the same Ni isotopic composition as the terrestrial standard within the uncertainties of our study [2], consistent with their age and their Fe/Ni ratios and in agreement with data by Cook et al. [3,4]. In contrast, Bizzarro et al. measured deficits in $^{60}Ni^*$ and $^{62}Ni$ in 7 iron meteorites, interpreted them as nucleosynthetic anomalies, and concluded that irons formed in the absence of $^{60}Fe$. This in turn would suggest a late injection of $^{60}Fe$ into the early solar system [5], which is in contradiction to our results on CAIs [6]. It is, however, important to note that Bizzarro et al.'s Ni isotope ratios are not resolvable from the terrestrial standard if our more conservative 2SD uncertainty is taken into account.

**CAIs:** Ca-Al-rich inclusions are the oldest condensates of the solar system. It may therefore be possible to deduce the initial $^{60}Fe/^{56}Fe$ ratio of the solar system from their Ni isotope composition,. The initial $^{60}Fe$ abundance provides constraints on the stellar environment surrounding the nascent Sun and determines whether $^{60}Fe$ was an important heat source for planetary melting. Most CAIs display nucleosynthetic anomalies for Ni [6,7]. However, nickel-60* anomalies are also observed in CAIs that do not carry nucleosynthetic anomalies: some live $^{60}Fe$ was present in the inclusions at the time they formed but the data are inconsistent with a simple undisturbed system [6]. An internal isochron for an Allende CAI provides a lower limit of 4.2_$10^{-7}$ for the initial $^{60}Fe/^{56}Fe$ of the solar system, but this CAI contains secondary minerals indicating that the Fe-Ni system most likely underwent re-equilibration. An initial value higher than 1.8_$10^{-6}$ is inferred from two CAIs with no nucleosynthetic anomalies and characterized by the same $^{26}Al/^{27}Al$

initial ratio. This estimate is consistent with results of nucleosynthetic modeling that predicts the observed correlated $^{60}$Fe, $^{62}$Ni, and $^{96}$Zr nucleosynthetic anomalies in CAIs [6]. It also agrees with a recent estimate based on chondrites (Wadhwa, personal communication), but is significantly higher than the value deduced from troilite in primitive chondrites [8]. The exact initial $^{60}$Fe/$^{56}$Fe thus remains poorly known.

**Iron meteorites:** Iron meteorites show no resolvable anomalies in $^{60}$Ni*; magmatic and non-magmatic irons cannot be distinguished from each other. Given their Fe/Ni ratios, some irons may have crystallized as early as 0.4 Myrs after the beginning of the solar system, whereas others probably cooled through the closure temperature of the Fe-Ni system much later [2]. This is compatible with the very old $^{182}$Hf/$^{182}$W age of irons, but no precise chronological conclusion can be drawn due to the uncertainties and to the small range of Fe/Ni ratios among irons.

**Eucrites and angrites:** All eucrite samples are characterized by well-resolved $^{60}$Ni* excesses, indicating that live $^{60}$Fe was present at the time the core formed on their parent body and also at the time the eucrites crystallized [9, 10]. Based on $^{182}$Hf-$^{182}$W chronology, partial melting of the mantle took place in the eucrite parent body about 4 Myrs after the beginning of the solar system. A back-calculation gives an initial $^{60}$Fe/$^{56}$Fe of about 4_10$^{-6}$, much higher than the other estimates. This may be due to a complex history of Fe and/or Ni redistribution in eucrites as these two elements are highly mobile.

The results presented above confirm that the Fe-Ni isotopic system is easily disturbed. To determine whether it can be used as a reliable chronometer in, at least, the best preserved samples, $^{60}$Fe-$^{60}$Ni data must be intercalibrated with other chronometers and/or an absolute timescale. Angrites are critical samples for this purpose because they formed very early in the solar system, are differentiated, and cooled quickly. Two angrites were selected here: Sahara 99555, a pristine quenched angrite in which the Fe-Ni system may have remained undisturbed, and North West Africa 2999. Markowski et al. [11] recently confirmed with W isotopes that NWA belongs to the group of “slowly cooled” angrites. The Ni isotopic composition of different mineral fractions from NWA 2999 varies between –0.04±0.20 and 0.03±0.26 (2SD) with an Fe/Ni ratio ranging from 15.9 to 58.9. The slope of a possible isochron is shallower than 3.75_10$^{-8}$, which indicates that the Fe-Ni system in NWA 2999 closed more than 8.3 Myrs after the beginning of the solar system. This is fully compatible with recent $^{182}$Hf-$^{182}$W data for this meteorite [11]. SAH 99555 formed earlier: 4562-4563 Myrs ago. Excesses of +12 to +40 $\varepsilon_{60}$ thus would be expected, assuming that core formation on the parent body took place 2.5 Myrs after the beginning of the solar system. This is not observed. No fraction (mineral separate or size fraction) yields a resolvable excess of $^{60}$Ni*, indicating that there was no or only very little live $^{60}$Fe present when SAH 99555 formed. These results conflict with our data obtained for CAIs but also for eucrites. In addition, a time interval of 13.2 Myrs is inferred between CAIs and SAH 99555 if one considers the best-fit line as an isochron, which is inconsistent with $^{26}$Al-$^{26}$Mg, $^{53}$Mn-$^{53}$Cr, $^{182}$Hf-$^{182}$W, and Pb-Pb ages for this angrite. It may be that the Ni isotope data for SAH 99555 have no chronological meaning. Bizzarro et al. detected small $^{60}$Ni* and $^{62}$Ni deficits in angrites and interpreted them as nucleosynthetic anomalies [5]. Our current measurements for NWA 2999 do not confirm this observation: all Ni isotope ratios are identical to the terrestrial standard. SAH 99555 is not as rich in Ni as NWA 2999 and more measurements are required to ascertain the precise $^{61}$Ni/$^{58}$Ni and $^{62}$Ni/$^{58}$Ni ratios.

**Conclusions:** Some, but not all, CAIs show $^{60}$Ni* anomalies due to the in situ decay of $^{60}$Fe. Large excesses of radiogenic Ni also are detected in eucrites, but neither in iron meteorites nor in angrites. This is difficult to reconcile with data from other chronometers. In addition, different approaches yield different estimates for the initial $^{60}$Fe/$^{56}$Fe of the solar system. Altogether, these observations show that (1) the $^{60}$Fe-$^{60}$Ni system cannot yet be used as a ubiquitously reliable chronometer and (2) $^{60}$Fe was probably heterogeneously distributed in the early solar system.

**References:** [1] Quitté G. and Oberli F. (2006) *JAAS, 21,* 1249-1255. [2] Quitté G. et al. (2006) *EPSL, 242,* 16-25. [3] Cook D. et al. (2005a) *LPS XXXVI,* Abstract #1179. [4] Cook D. et al. (2005b) *MAPS, 40,* A33. [5] Bizzarro M. et al. (2006) *MAPS, 41,* Abstract #5217. [6] Quitté G. et al. (2007) *ApJ, 655,* 678-684. [7] Birck J.L. and Lugmair G.W. (1988) *EPSL, 90,* 131-143. [8] Mostefaoui S. et al. (2005) *ApJ, 625,* 271-277. [9] Shukolyukov A. and Lugmair G.W. (1993) *EPSL, 119,* 159-166. [10] Quitté G. et al. (2005) *LPS XXXVI,* Abstract #1827. [11] Markowski et al. (2006) *MAPS, 41,* Abstract #5195.