THE ISOTOPE GEOCHEMISTRY OF NICKEL IN CHONDRITES AND IRON METEORITES. F. Moynier, P. Té-louk, J. Blichert-Toft, and F. Albarède, Ecole Normale Supérieure de Lyon (46 allée d’Italie, 69364 Lyon cedex 7, France, albarede@ens-lyon.fr).

Introduction: Difficulties with chemical separation and mass spectrometry combined with little expectation of isotopic fractionation at high temperature left the stable isotope geochemistry of nickel unattended. The quest for \(^{60}\)Ni excess resulting from the decay of the radioactive nuclide \(^{60}\)Fe \((T_{1/2} = 1.5\, \text{My})\) was, however, successful but the small amount of data produced to date attests to the particularly difficult measurement by TIMS (Birck and Lugnair, 1988; Shukolyukov and Lugnair, 1993a,b) and SIMS (Kita et al., 2000). The potential of MC-ICPMS techniques is strong (Quitté et al., 2002) but has not been fully explored. This preliminary report describes evidence of mass-dependent fractionation of the stable Ni isotopes \(58\) (68.1‰), \(61\) (1.14‰), \(62\) (3.63‰), and \(64\) (0.93‰) as measured by MC-ICPMS in the metal phase of ordinary chondrites, in some of their coexisting silicate phases, and in iron meteorites.

Techniques: Metal was separated from the sample powders using a hand magnet and dissolved in HCl. Silicates were hand-picked and dissolved in HF. Ni was purified using a combination of anion and cation-exchange chemistry. The samples were run on the Plasma 54 MC-ICPMS of Lyon. Cu was used to correct for the instrumental mass bias. Isobaric interferences, notably ArNe at mass 60, remain below 10^-11 A and were corrected using blank solutions. The sample external reproducibility on each ratio is 50 ppm (this figure probably overestimates the error bar on the 60/58 ratio). This precision can be reduced by pooling several runs. We have not been able, so far, to identify isotopic variations among high-temperature terrestrial minerals: we therefore report all the results with respect to the Aesar ICP MS Ni standard solution.

Results: The data are reported in delta permil notation per mass difference unit, for instance, \(\delta^{58\text{Ni}}/\text{Ni/3 or }\delta^{62/58\text{Ni}}/4\text{, which ensures that mass-dependent fractionation leaves the point on a 1:1 straight line. The metal phase of ordinary (H, L, and LL) chondrites with different degrees of metamorphic alteration (3 to 6) and three iron meteorites have been analyzed. Three silicate phases were also analyzed but the sample was to small (10^-11 A total Ni) for a precise signal to be obtained at masses 61, 62, and 64. Fig. 1 (bottom) shows that mass fractionation on \(^{60}\text{Ni/58}\text{Ni}\) and \(^{62}\text{Ni/58}\text{Ni}\) is mass-dependent. Similar results were obtained for the \(^{60}\text{Ni/58}\text{Ni}\) ratio. When ratios are plotted vs \(^{62}\text{Ni/58}\text{Ni}\), the scatter of the \(^{60}\text{Ni/58}\text{Ni}\) data about the mass discrimination line is larger in spite of their better precision. This scatter attests to the presence of about 150 ppm variations in the radiogenic \(^{60}\text{Ni/58}\text{Ni}\) ratio among the different samples. The largest \(^{60}\text{Ni}\) excess is found in the metal phases of Forest Vale and Estacado, the largest deficit in Tieschitz. The range seems independent of the petrographic group. Two iron meteorites (Casas Grandes and Canyon Diablo) show a significant \(^{60}\text{Ni}\) excess.

The variations of the stable ratios (excluding \(^{60}\text{Ni/58}\text{Ni}\)) are small but measurable. The range of mass fractionation per mass difference unit in the metal phases of chondrites extends over 300 ppm (0.3 delta permil or 3 epsilon units). The mean value seems related to the petrographic type: +0.05 ‰ for LL, +0.15 ‰ for L, and +0.25 ‰ for H. The iron meteorite samples seem to be heavy (+0.15 ‰ to +0.28 ‰). The lowermost values (-0.03 ‰) are obtained for Mezö-Madaras which is currently re-run to check that the Ni of this sample was not isotopically fractionated during its purification. Although the silicate samples run so far are few, they are definitely lying on the negative side (-0.014 ‰ to -0.72 ‰). We have not found so far any compelling relationship between the Ni isotope composition and the degree of metamorphic transformation.

Discussion: Clear evidence is obtained that, as for Fe (Zhu et al., 2001), the stable isotopes of nickel fractionate out of a common metal pool. When both the silicate and metal phases are considered, the total range of Ni isotopic variations in the Solar System must be in excess of 1 ‰. Since most of the Ni inventory of the sample is hosted in the metal phase, the range of isotopic variations in the bulk material is rather 0.3 ‰. The seemingly systematic enrichment of the heavier Ni isotopes observed in the metal phase from the LL to the H suggests that volatility controls isotopic variations. There are not enough Ni isotope data on silicates at this point to draw a strong conclusion, but if the metal-silicate fractionation observed on the present samples holds for the Earth, the Ni isotope composition of the Earth’s core—and of the Earth as a whole—may be close to that of H chondrites and possibly heavier (E?)

Since most of the Earth’s Ni inventory is in the core, the presence of \(^{60}\text{Ni}\) excesses and deficits in the metal phase of chondrites with respect to terrestrial silicates (our standard) can be used to date the onset of the Earth’s core segregation. On average, no strong \(^{60}\text{Ni}\) anomaly is detected, which may signal that core
segregation only started after most of the initial $^{60}$Fe had decayed away (a few My).


Figure 1: Ni isotopic data on the metal phases of H, L, and LL chondrites and on iron meteorites. The values are in delta ‰ divided by the difference between the masses of the numerator and denominator isotopes, so that the diagonal line represents a normalized mass-dependent linear fractionation law. Bottom: the stable isotopes $^{54}$Ni, $^{58}$Ni, and $^{62}$Ni follow a mass-dependent fractionation law. Top: the spread around the mass fractionation line is greater than in the figure below: this shows that small amounts of excess/deficit of radiogenic $^{60}$Ni is present.