S. Mostefaoui et al. / New Astronomy Reviews xx (2003) pp-pp

Evidence for live ^{60}Fe in meteorites

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Abstract

We report a preliminary *in situ* finding of ^{60}Ni isotopic anomalies, attributed to the decay of short-lived ^{60}Fe (half-life 1.5 Ma), in Fe-sulfides and pyroxene in the Chervony Kut eucrite and in the Semarkona (LL3.0) ordinary chondrite using the new state-of-the-art NanoSIMS technique at MPI in Mainz. In Semarkona, troilites (FeS) show ^{60}Ni excesses of up to 46 ± 15‰ (2σ). A positive correlation between ^{60}Ni excesses and ^{56}Fe/^{58}Ni ratios provides evidence for live ^{60}Fe in the early solar system. The inferred ^{60}Fe/^{56}Fe ratio of $(7.3 \pm 2.6)\times10^{-7}$ is much higher than previously estimated for the initial solar system value. In terms of a chronological interpretation, this corresponds to one half-life of ^{60}Fe. In Chervony Kut, two pyrrhotite $\{Fe_{(1-x)}S\}$ types (abundant Pyr-1 crystals and rare Pyr-2 veins) and pyroxene $\{(Mg,Fe)SiO_3\}$ grains show ^{60}Ni excesses. The delta-^{60}Ni varies from 22 ± 16‰ (2σ) in Pyr-1 to an extremely high value of 1775 ± 250 ‰ in Pyr-2 veins. No clear correlation is seen between the ^{60}Ni excess and the Fe/Ni ratio, which does not allow a plausible explanation for the extreme excess encountered in Pyr-2. More meticulous analyses are underway in order to confirm this preliminary finding.

Keywords: meteorites, Solar system, extinct radionuclides, isotope abundance anomalies, planetary differentiation.
PACS: 96.50.Mt, 26.45.+h, 07.75.+h,26.30.+k

1. Introduction

The nucleosynthetic production of short lived radionuclides and their rapid injection into the nascent solar system offer information about the time scale of events that occurred during the early history of the solar system. Meteorites represent the oldest solar system matter that we have access to. Their study reveals evidence for the existence and decay of these radionuclides through the detection of anomalies in their daughter isotopes.

Chronometers based on short lived nuclides, such as ^{26}Al, ^{53}Mn, ^{60}Fe, ^{107}Pd, ^{129}I, and ^{146}Sm, were extensively studied during the last decades (Lee et al., 1976; Kelly and Wasserburg, 1978; Lugmair et al., 1983; Birck and Allegre, 1985; Swindle et al., 1988; Shukolyukov and Lugmair, 1993). Some of these nuclides were shown to afford chronological resolutions on the order of a few million years (My) to less than one My (e.g. Lugmair and Shukolyukov, 1998; Mostefaoui et al., 2002). Some short-lived radionuclides were also considered as heat sources inducing planetary differentiation (Urey, 1955; Hutcheon and Hutchison, 1989; Shukolyukov and Lugmair, 1993; Shukolyukov and Lugmair, 1997). It can be safely stated that ^{26}Al served as a heat source for early planetary melting and differentiation. Considering the short half-life of ^{60}Fe and the high iron content of meteorites, ^{60}Fe is another potential heat source for planetary melting and differentiation. However, due to a scarcity of phases with high Fe/Ni ratios and analytical limitations and difficulties, ^{60}Fe was not explored as extensively as ^{26}Al. The first resolvable excess of ^{60}Ni (the daughter product of ^{60}Fe) was shown to exist in the Chervony Kut eucrite by Shukolyukov and Lugmair (1993). A positive correlation between ^{60}Ni excesses and Fe/Ni ratios was taken as evidence that live ^{60}Fe had existed in the early solar system. Since then, two questions have been addressed: Can the Fe-Ni system be used as a chronometer and can ^{60}Fe be a heat source for planetary melting and differentiation, and if so, to what extent? In a quest to help answer these questions, we report here on new and preliminary results on ^{60}Ni isotopic anomalies in two meteorites using the new state-of-the-art technique of the NanoSIMS.

2. Dating events using extinct radioactivity

In this section we briefly review the principle of radiochronology utilizing extinct radionuclides to advance the appreciation of the data below and their implications. The parent radioactive nucleus decays with its characteristic half-life to a stable daughter nucleus. This decay follows the well known expression $P_t = P_0\exp(-\lambda t)$, where λ is the decay constant and P_i are the number of atoms of the parent nucleus at the time of formation of a closed system (t = 0) and after a decay interval t. If D_0 is the number of atoms of the daughter nucleus already present at t = 0, then the simple expression $P_0 + D_0 = P_t + D_t$ must be valid. This forms the basis for radiochronology.

In practice, isotope ratios can always be measured with much higher precision than the number of atoms of a single isotope. Thus, stable isotopes of the parent and daughter elements are used in the denominator of the isotope ratios. In the case of ^{60}Fe, which decays to ^{60}Ni by β emission with a half-life of 1.5 My, the ^{60}Ni now present in any natural object is the sum of the initial ^{60}Ni present in this object and the ^{60}Ni due to the decay of ^{60}Fe. A simple transformation of the above equation then results in the more useful expression:

$$\left(\frac{^{60}\mathrm{Ni}}{^{58}\mathrm{Ni}}\right)_{\mathrm{Pres}} = \left(\frac{^{60}\mathrm{Ni}}{^{58}\mathrm{Ni}}\right)_{\mathrm{Init}} + \left(\frac{^{60}\mathrm{Fe}}{^{56}\mathrm{Fe}}\right)_{\mathrm{Init}} * \left(\frac{^{56}\mathrm{Fe}}{^{58}\mathrm{Ni}}\right)_{\mathrm{Pres}}$$

where ‘Pres’ stands for present value and ‘Init’ for the initial value at time t = 0. This equation is that of a straight line.

For the initial ^{60}Ni/^{58}Ni ratio, the value of 0.38218 (Birck and Lugmair, 1988) can be used and the present ^{60}Ni/^{58}Ni and ^{56}Fe/^{58}Ni ratios are measurable (in our case with the NanoSIMS). The inferred initial ^{60}Fe/^{56}Fe ratio of the object is given by the ‘slope’ in the above equation. It can be determined by the measurement of several mineral phases having different chemical compositions (*i.e.* Fe/Ni ratios). Inversely, the existence of a correlation between the measured ^{60}Ni/^{58}Ni and ^{56}Fe/^{58}Ni ratios is a clear indication for life ^{60}Fe in the measured phases. Generally, the two above quantities are presented in a diagram, with ^{60}Ni/^{58}Ni sometimes denoted in δ-units (see Fig. 2), which expresses the deviation in permil from the initial ratio. Once ^{60}Fe/^{56}Fe is determined with an acceptable precision, the time difference between the closures of two different systems (relative age of a rock or of an assemblage of phases to another such system) can be expressed as:

$$\Delta t = \lambda * \ln[(^{60}\mathrm{Fe}/^{56}\mathrm{Fe})_1/ (^{60}\mathrm{Fe}/^{56}\mathrm{Fe})_2]$$

where the subscripts denoting the two different systems and λ the decay constant of ^{60}Fe.

3. Samples

In this work we studied Fe-Ni isotopes in two meteorites using the NanoSIMS. These two meteorites are the Chrevony Kut eucrite and the Semarkona LL3 ordinary chondrite. These two meteorites were chosen because Chervony Kut is known to have excess ^{60}Ni, and because the study of its material can give information on planetary differentiation (Gooding et al., 1979). However, the interpretation of previous data obtained on mineral separates and on a macroscopic scale, was complicated by shock-metamorphic effects. In contrast, Semarkona is a very primitive chondrite where the disturbance of the Fe-Ni system during parent body processing is expected to be minimal. This may allow us to obtain a better estimate of the initial solar system ^{60}Fe abundance.

In Semarkona, despite the complex petrography, we can clearly distinguish between metal-associated troilite and metal-free troilite (Fig. 1).

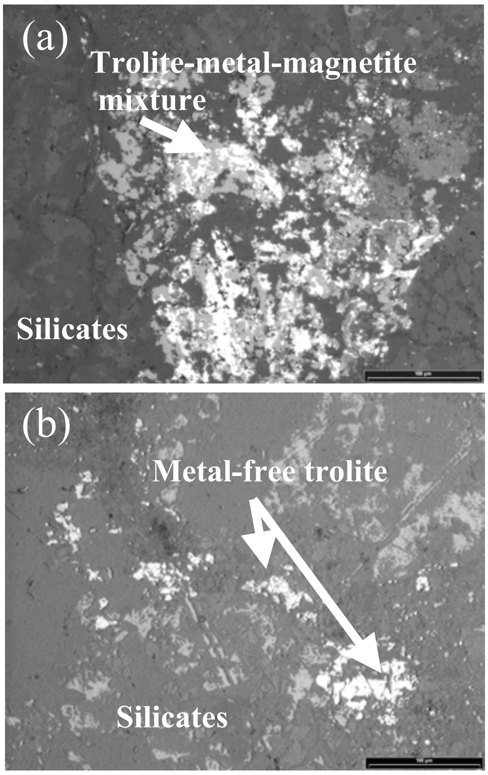

Fig 1. Optical microscope images of (a) a troilite-metal-magnetite assemblage and (b) metal-free troilite aggregates in Semarkona matrix. The scale bars are 100μm.

In metal-associated troilite, the troilite is jointly attached or in contact with metal and magnetite (Fig. 1a). The association of the three phases is common in Semarkona, and assemblages of up to several hundreds of microns can be found. Diffusion in such assemblages is expected from the troilite to the metal phases, thus disturbing the ^{60}Fe-^{60}Ni radiogenic system. The metal-free troilite is thought to be primary and no diffusive interaction with metal could have occurred. This troilite can have high Fe/Ni ratios, making it our principal target for the search of possible excesses in ^{60}Ni. Our NanoSIMS measurements of the troilite were made on three aggregates of this type.

The petrography of the Chervony Kut eucrite is much simpler than that of Semarkona. In Chervony Kut, we found two types of pyrrhotite $\{Fe_{(1-x)}S\}$. The first type (Pyr-1) is abundant, and the grains are variable in size (from 1 to 100μm) and round in shape. The second type of pyrrhotite (Pyr-2) is present as veins in a shock-melt pocket. Pyr-2 was found in only one location. The phases we analyzed in Chervony Kut are the two pyrrhotite types and pyroxene.

4. Technique

The Fe and Ni isotopes were measured with the Cameca NanoSIMS-50 ion microprobe. The NanoSIMS is a new generation secondary ion mass spectrometer, installed 2 years ago at the Max-Planck Institute for Chemistry at Mainz. (see also the contributions by Hoppe et al. and Ott et al. in these proceedings). Measuring conditions of the NanoSIMS were as follows: Positive secondary ions of ^{54}Fe, ^{60}Ni, and ^{62}Ni were measured in a multi-detection mode at a mass resolution m/Δm of ~4000. Using high primary current conditions (~ 1 nA on the sample surface), the primary ion beam of O^- was focused into spots of 1 to 5 μm in size on the sample. The instrumental mass fractionation for ^{60}Ni/^{62}Ni ratios was corrected using external standards: For Semarkona, we used Ni-rich phases found in the sample itself in the vicinity of the selected troilites, and for Chervony Kut a synthetic Fe-Ni-rich standard.

5. Results

In Semarkona troilite, the ^{56}Fe/^{58}Ni ratios range up to ~10^4 (Fig. 2). ^{60}Ni excesses were found in the three measured troilite aggregates, with a maximum δ^{60}Ni of 46±15 (2σ).

Figure 2 displays the relative excesses expressed in δ-units as a function of the ^{56}Fe/^{58}Ni ratio of Semarkona troilite.

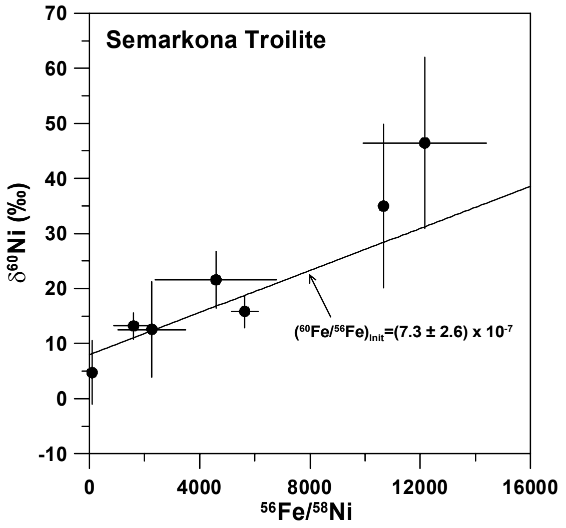

Fig 2. δ^{60}Ni as a function of ^{56}Fe/^{58}Ni for three troilite aggregates in the Semarkona matrix. ^{56}Fe and ^{58}Ni are calculated from measured ^{54}Fe and ^{60}Ni. Errors are 2σ.

Each data point comprises the results of one to six measurements on the same troilite grain. The diagram shows a common trend for the three troilite aggregates - a clear correlation is seen between ^{60}Ni excesses and ^{56}Fe/^{58}Ni ratios. The best-fit line through the data yields an inferred ^{60}Fe/^{56}Fe ratio of (7.3 ± 2.6) x 10^{-7} (2σ error). The non-zero intercept of this line is probably due to matrix effects and needs further investigation.

In Chervony Kut, the ^{56}Fe/^{58}Ni ratios are between ~2x10^4 and ~1x10^5 in pyrrhotite and up to around 5x10^5 in pyroxene. These high ratios are not surprising, since Chervony Kut is known to have a bulk Ni content of only a few ppm. ^{60}Ni excesses are found in pyrrhotite and also in pyroxene. The δ^{60}Ni varies from 22±16‰ (2σ) in Pyr-1 to an extremely high value of 1775 ± 250 ‰ in Pyr-2 veins. The data for Pyr-2 are the result of four measurements taken from two different grains.

For Chervony Kut, there is no clear correlation between ^{60}Ni excesses and Fe/Ni ratios. Nevertheless, a forced fit-line through the origin using the Pyr-1 and Pyroxene data gives a ^{60}Fe/^{56}Fe ratio of (7.8 ± 4.2) x 10^{-8}, which is significantly higher than that of the bulk Chervony Kut samples by Shukolyukov and Lugmair (1993). Interestingly, however, the value of 5‰ excess obtained by Shukolyukov and Lugmair (1993) on a "troilite" grain falls exactly on this line.

6. Discussion

The correlation in Fig. 2 gives evidence for live ^{60}Fe in Semarkona. From this measurement the resulting ^{60}Fe/^{56}Fe ratio of ~7x10^{-7} is much higher than previously estimated for the initial solar system value by Shukolyukov and Lugmair (1993) and approaches that inferred from a Calcium-Aluminum-rich Inclusion (CAI) from the Allende meteorite (1.6x10^{-6}; Birck and Lugmair, 1988). In terms of a chronological interpretation, assuming a homogeneous distribution of ^{60}Fe in the early solar system, this preliminary value from the troilite in Semarkona corresponds to a difference of only about one half-life of ^{60}Fe. This would indicate that the Semarkona troilite was formed ~1My after CAIs, believed to be the first mm-sized objects formed in the solar nebula.

The presence of ^{60}Fe in the Semarkona materials shows that ^{60}Fe was alive at the birth of the solar system. ^{60}Fe cannot be produced by irradiation by energetic particles from the early sun (Lee et al., 1998). Also, the ^{60}Fe/^{56}Fe ratio reported in this study is too high compared to the abundance reported for continuous galactic evolution (Wasserburg et al., 1996). The most probable source for such a high level of ^{60}Fe would be a supernova explosion (Wasserburg et al., 1998). Considering the short half-life of ^{60}Fe, the supernova explosion most likely occurred shortly before or at the birth of the solar system. This would also support the scenario of a supernova trigger being responsible for the formation of the solar system (Cameron et al., 1995).

A ratio of ~7x10^{-7} is high, and the question is, can ^{60}Fe produce sufficient heat to induce planetary melting? Recently, Yoshino et al. (2003) constructed a model describing the production of heat based on the decay of ^{26}Al and ^{60}Fe in asteroids of chemical compositions similar to carbonaceous chondrites and with a radius of more than 30 km. Based on their Fig. 2, if a low initial ^{26}Al/^{27}Al ratio of 7.7x10^{-6} is taken, an initial ^{60}Fe/^{56}Fe ratio between 2x10^{-7} and 2x10^{-6} would be adequate to increase the temperature to levels that leads to melting of rocks within the first million years of solar system existence. Our new value thus strongly suggests that ^{60}Fe was an important heat source contributing to planetary differentiation.

The data for Chervony Kut are much more complex, although the petrography is much simpler as compared to Semarkona. The ^{60}Ni excess in the Pyr-2 veins is surprisingly high. This is the highest excess ever reported for ^{60}Ni. We know that Chervony Kut is a differentiated meteorite. This is seen not only from the petrography but also from the very low bulk Ni content of the meteorite, suggesting that there was complete melting and segregation of metal towards the core of the parent asteroid. However, at present we have no satisfactory explanation for the survival of the Pyr-2 lithology. More measurements will certainly be needed to understand the early evolution of the Chervony Kut parent asteroid.

7. Conclusions

From these preliminary results we can conclude that: (1) Metal-free troilites in Semarkona (LL3.0) show resolvable ^{60}Ni-excesses, with $\delta^{60}\mathrm{Ni}_{\mathrm{max}} < 60‰$. (2) The correlation of δ^{60}Ni with Fe/Ni indicates live ^{60}Fe in the early solar system, with $^{60}\mathrm{Fe}/^{56}\mathrm{Fe}_{\mathrm{init}} \geq 7\mathrm{x}10^{-7}$. (3) The troilites in Semarkona were probably formed ~1My after CAIs (assuming a homogeneous ^{60}Fe distribution within the solar system, with an initial ^{60}Fe/^{56}Fe ratio of 1.6x10^{-6}). (4) ^{60}Fe was produced in a supernova and injected into the solar system shortly before or at its birth. (5) ^{60}Fe is an important heat source for planetary melting and differentiation. (6) Resolvable ^{60}Ni-excesses are detected in pyroxene and in two types of pyrrhotite (Pyr-1 & Pyr-2) in the Chervony Kut eucrite. (7) Pyr-2 veins show an extreme excess in ^{60}Ni with δ^{60}Ni of ~+1775‰.

Acknowledgements: We are grateful to G. J. MacPherson and the Russian Academy of Science for the loan of the Semarkona and Chervony Kut samples. We thank E. Gröner for technical assistance on the NanoSIMS.

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