NICKEL NUCLEOSYNTHETIC ANOMALIES IN LEACHATES OF CARBONACEOUS CHONDRITES.

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Introduction: The numerous nucleosynthetic isotopic anomalies found in refractory inclusions, in presolar grains, and in special phases carried by chondrites suggest that the matter constituting the solar system results from a mixing between different sources. Recently, Ni nucleosynthetic anomalies have been found in various meteorites, including iron meteorites, ureilites, chondrites [e.g. 1-3], and building material of meteorites has been shown to come from at least 3 isotopically distinct reservoirs for Ni [3]. The carrier phases of the Ni isotope anomalies have however not been fully identified yet, despite a first study based on iron meteorites and chondrites [2].

Carbonaceous chondrites can be used to identify the nucleosynthetic sources of solar system material and to study stellar nucleosynthesis and formation of the solar system. Stepwise dissolution of carbonaceous chondrites is a powerful tool to characterize the fine-scale isotope heterogeneities of the solar system and to identify the presolar carrier phases of isotope anomalies. Acid leachates of these meteorites already display nucleosynthetic anomalies for Cr, K, Mo, Ba, Zr, Os [4-9].

Here we report Ni isotope analyses of leachates for the three primitive carbonaceous chondrites Allende, Murchison, and Orgueil with the aim of resolving the different nucleosynthetic components. Nickel is a suitable element for this purpose as ^{61}Ni is overproduced by s-process when excesses of ^{62}Ni and ^{64}Ni witness nucleosynthesis in a neutron-rich environment. Besides, possible correlations between different isotopes or different elements in the same leachates potentially bring stronger and more precise constraints on the astrophysical setting where the nuclides have been produced. As an example, correlations between ^{60}Fe, ^{62}Ni and ^{96}Zr have already been observed in CAIs [10] and ^{62}Ni and ^{54}Cr anomalies seem to be correlated in several types of meteorites [1].

Analytical procedure: Stepwise dissolutions similar to those previously used in several other studies [e.g. 4,7] have been applied for Ni to facilitate the comparison of the isotopic results. Powdered whole rock samples of Allende (CV3), and Murchison (CM2) were sequentially digested with reagents of increasing acid strength following the sequence described in Table 1:

Step	Reagent	Procedure
1	2.5% HAc	30 min, RT
2	50% HAc	1 day, RT
3	4N HNO3	5 days, RT
4	6N HCl	5 days, RT
5	6N HCl	1 day, 36°C
6	6N HCl	1 day, 80°C
7	13.5N HF – 3N HCl	4 days, 100°C
8	HF-HCl-HNO_3	4 days, 145°C

Table 1: leaching procedure for Allende and Murchison.

The powdered Orgueil (C1) sample was originally dedicated to the search of ^{58}Fe anomalies and a slightly different leaching procedure has been applied: steps 1 and 2 using acetic acid have been combined in a single step, while step 2 for Orgueil is comparable to step 3 of Allende and Murchison even if the leaching lasted for two more days.

Step	Reagent	Procedure
1	17N CH_3COOH	1 day, RT
2	4N HNO_3	7 days, RT
3	6N HCl	5 days, RT
4	6N HCl	1 day, 35°C
5	6N HCl	1 day, 80°C

Table 2: leaching procedure for Orgueil.

Nickel has been separated from the matrix elements and purified using ion exchange resins. Nickel isotopes have then been measured using a MC-ICPMS instrument at ENS Lyon. Thanks to its large mass dispersion, instrument Nu 1700 permits to collect simultaneously all Ni isotopes as well as ^{56}Fe, ^{57}Fe and ^{66}Zn to correct for the isobaric interferences on masses 58 and 64. Nu 1700 also has high mass resolution capabilities so that argide interferences can be fully resolved using a mass resolution of 2600. The external reproducibility (2*SD) achieved for a standard solution is about 15ppm for ^{60}Ni/^{58}Ni, 30ppm for ^{61}Ni/^{58}Ni and 100ppm for ^{64}Ni/^{58}Ni when the ratios are internally normalised to ^{62}Ni/^{58}Ni.

Results and Discussion: Most of the Ni is dissolved by concentrated acetic acid and nitric acid confirming that metal is a major carrier phase for Ni. In Orgueil, Ni is not isotopically uniform among its various host phases. The apparent ^{61}Ni-excesses translate into ^{62}Ni-deficits when the ^{61}Ni/^{58}Ni ratio is used instead of ^{62}Ni/^{58}Ni for internal normalisation. All fractions have a similar isotopic pattern but the ^{60}Ni-deficit increases with increasing acid strength. The initial release fractions are deficient in ^{62}Ni by 9ppm and the subsequent fractions by 40-45ppm. As ^{58}Fe is the most neutron-rich Fe isotope, a correlation is expected with the neutron-rich Ni isotopes that also belong to the iron peak elements and are co-produced in the same nucleosynthetic environment, but no ^{58}Fe anomalies have been observed in Orgueil within uncertainty [11]. Hints towards negative $\varepsilon(^{58}\mathrm{Fe}/^{54}\mathrm{Fe})$ values nevertheless exist but the anomalies are not resolvable from the standard. In fact, Fe and Ni anomalies may be decoupled as Fe is much more abundant than Ni in the protoplanetary disk so that it is less easily "contaminated" by isotopically anomalous carrier phases, due to dilution effects. A mass balance calculation for Ni in Orgueil does not yield the composition of the bulk sample. This is most likely due to the truncated leaching procedure. Indeed, the last release fractions using HF were not available for Ni isotope measurements while these leachates show an isotopic signature complementary to those of the previous steps in Allende and Murchison.

In contrast to Orgueil, no Ni isotopic anomaly can be clearly resolved from the terrestrial standard for most fractions of Allende and Murchison with the current analytical precision. These meteorites may nonetheless contain some phases bearing a distinct nucleosynthetic signature. Two complementary nucleosynthetic components may be distinguished: one enriched in ^{61}Ni (s-process product) or depleted in ^{62}Ni, and another depleted in the s-process nuclide or enriched in ^{62}Ni. Note that these first results need confirmation with reduced uncertainties on the data.

Iron-60, though not alive anymore, was at the beginning of the solar system the most neutron-rich isotope of Fe. Nickel data obtained on CAIs have demonstrated that ^{60}Fe and ^{62}Ni have been co-produced in the stellar source [10]. Present ^{60}Ni/^{58}Ni and ^{62}Ni/^{58}Ni values measured in the different release fractions also plot on a positive trend, indicating that ^{60}Ni is the decay product of live ^{60}Fe at the beginning of the solar system. However, contrary to what is observed in CAIs, there is a lot a scatter around the ε_{60}-ε_{62} correlation, which may be consistent with a heterogeneous distribution of ^{60}Fe in the early solar system.

Tungsten isotope analyses of the same leachates are in progress and will be used to test the nucleosynthesis model of Meyer and Adams suggesting co-production of ^{62}Ni and ^{182}Hf in the same stellar environment.

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