CORRELATED NEUTRON RICH Ni ISOTOPE ANOMALIES IN CHONDRITIC AND IRON METEORITES.

R. C. J. Steele[1]*, T. Elliott[1], C. D. Coath[1], M. Regelous[2], S. S. Russell[3]. [1]Bristol Isotope Group, Dept. Earth Sciences, University of Bristol, UK. *r.steele@bristol.ac.uk. [2]GeoZentrum Nordbayern, Universität Erlangen-Nürnberg. [3] Natural History Museum, London, UK.

Introduction: The Ni isotopic system is of particular interest in the study early solar system processes, and the sources of the material from which the solar system is made. The five stable isotopes of Ni have input from different nucleosynthetic environments enabling investigation of the stellar origins of this material. When the effects of early solar system processes that produce mass-dependent fractionation are removed, isotopic anomalies describe mixtures of these sources. Tracing isotopic anomalies in systems such as Ni between different meteorite groups can yield information about the degree of heterogeneity in the early solar system and the processes required to retain or produce this heterogeneity. Furthermore, ^{60}Ni has input from the short-lived ^{60}Fe ($t_{1/2}$ = 2.62 ± 0.04 Ma [1]). The initial abundance of live ^{60}Fe is of particular interest to the study of the early solar system as a heat source for planetary melting and as a high resolution chronometer.

Debate surrounds the existence of Ni isotope anomalies in bulk samples. While [2] and [3] reported anomalies in ε^{62}Ni and ε^{60}Ni other studies [e.g. 4, 5, 6, 7] find no evidence for resolvable anomalies in either ratio. One point of agreement is that if anomalies exist, they are small ($<1\varepsilon$) so their resolution requires high precision analyses ($< 0.15\varepsilon$ 2 SD) which increases the analytical challenge associated with their measurement.

The debate can be extended by examining the most neutron rich stable Ni isotope, ^{64}Ni. Nucleosynthetic theory predicts ^{64}Ni will exhibit larger anomalies than ^{62}Ni [e.g. 8, 9, 10, 11] for the type Ia supernovae (SNIa) events hypothesized to produce positive ε^{62}Ni anomalies. This was confirmed by early measurement in CAIs [12], but anomalies in bulk meteorites are smaller and harder to detect. Moreover, ^{64}Ni is the least abundant and most interfered Ni isotope making its measurement more problematic and requiring careful control on accuracy.

Analyses: We have built on the analytical procedure described by [2] to include analysis of ^{64}Ni. Separation of Ni from matrix elements is achieved using a four column procedure, utilizing the Ni specific complexing agent dimethylglyoxime in the first step. The Zn is removed by a final AG-MP-1 column and is reduced to blank levels ($Zn/Ni < 1.2 \times 10^{-5}$). Analyses were made on a Thermo-Finnigan Neptune MC-ICP-MS in medium resolution ($M/\Delta M > 6000$, where ΔM is peak width from 5-95% of peak height). Use of two Faraday cups connected to amplifiers with $10^{10}\Omega$ feedback resistors for measuring the most abundant isotopes, ^{58}Ni and ^{60}Ni, allows for beam intensities of up to 4 nA of ^{58}Ni. Sample measurements are bracketed by measurements of the NIST SRM 986 Ni isotopic standard. Standard and sample measurements are internally normalized to ^{58}Ni/^{61}Ni using the exponential mass bias law. A second normalization is applied to the samples by calculating epsilon (ε) units (parts per ten thousand difference) from interpolated ratios of NIST SRM 986. The 2s.e. (standard error) of our measurements, based on at least four repeats of each sample, are typically 0.02 ε^{60}Ni, 0.03 ε^{62}Ni and 0.08 ε^{64}Ni.

Results: We find larger variation in ε^{64}Ni ($\sim 0.90\varepsilon$) than previously noted for ε^{62}Ni [2] and a strong positive correlation between ε^{62}Ni and ε^{64}Ni. There is no overall correlation between ε^{60}Ni and ε^{62}Ni. Our new ε^{60}Ni and ε^{62}Ni data show no significant differences from [2] for chondrites or iron meteorites. This includes reanalysis of the same solutions as [2] and new dissolutions of meteorites from the same groups. The carbonaceous chondrites (CCs) have positive ε^{62}Ni ($\sim +0.15\varepsilon$) and ε^{64}Ni ($\sim +0.40\varepsilon$) and negative ε^{60}Ni ($\sim -0.10\varepsilon$). This is in contrast to the ordinary chondrites (OCs), which have negative ε^{62}Ni ($\sim -0.08\varepsilon$) and ε^{64}Ni ($\sim -0.20\varepsilon$) coupled with negative ε^{60}Ni ($\sim -0.05\varepsilon$). The enstatite chondrites (ECs) have ratios indistinguishable from Earth ($\sim 0.00\varepsilon$).

Interestingly, iron meteorites span approximately the same range as the chondrites. The main magmatic irons (IIAB, IIIAB and IVA) have ratios similar to the OCs. While the CO/CV/CM CCs have similar ratios to the IVB irons, and the CIs are more positive in both ε^{62}Ni and ε^{64}Ni.

Discussion: We have assessed, and eliminated, all perturbations to our data from: interferences on normalizing isotopes, residual instrumental mass bias and residual natural mass bias. We find no evidence that any of these process have significantly affected our data.

The controls on the accuracy and precision of ε^{62}Ni and ε^{64}Ni are different. The major challenge associated with the measurement of ε^{62}Ni is the requirement for high precision due to the small magnitude of anomalies ($\sim 0.4\varepsilon$). With larger anomalies ($\sim 0.9\varepsilon$) ε^{64}Ni is less affected by this, however the significant interference on ^{64}Ni from ^{64}Zn is a major factor controlling the accuracy of ε^{64}Ni measurements. Indeed the Zn interference has proved insurmountable in several previous studies [5, 7, 2]. In addition, the correlation of both ε^{62}Ni and ε^{64}Ni, with other neutron rich isotopes, eg ^{50}Ti or ^{54}Cr [13, 14], which is expected from nucleosynthetic theory, is further support of the validity of our data.

A recent study [6] found no resolvable anomalies in a range of meteoritic materials. We argue that the data from this study are not inconsistent with the data from [6] for two reasons. Firstly, the higher precision we achieve enables us to resolve the small anomalies present in the groups examined by [6]. Secondly, [6] did not analyse the groups which show the largest anomalies – the IVB irons and the CCs. In fact the data from [6] show the same, albeit barely resolvable, correlation which can be seen by plotting the data together with that from the present study.

From nucleosynthetic theory [e.g. 8, 9, 10, 11] the correlation between ε^{62}Ni and ε^{64}Ni can be interpreted as differential input SNIa material, preserved by incomplete mixing in the early solar system. This would also predict a positive correlation between ε^{60}Ni and ε^{62}Ni, unless all Fe had decoupled from Ni prior to significant decay of ^{60}Fe. However, there is no overall correlation between ε^{62}Ni and ε^{60}Ni. One hypothesis to explain this lack of correlation is heterogeneous distribution of live ^{60}Fe in the early solar system.

Conclusion: Mass-independent Ni isotope measurements in meteorites can provide a useful tool for investigating the nucleosynthetic source of the solar system and the processes and mechanisms by which they were mixed. Correlated ε^{62}Ni and ε^{64}Ni in bulk chondritic and iron meteorites suggests a variable contribution from a type Ia supernova component in the early solar system.

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