LARGE s-PROCESS AND MIRROR OSMIUM ISOTOPIC ANOMALIES WITHIN THE MURCHISON METEORITE.

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Introduction: Unequilibrated chondrites contain presolar grains with unique isotopic signatures reflecting the nucleosynthetic processes occurring in the stars in which they formed. The distribution of this ancient material, as deduced from isotopic variations among various classes of iron and chondritic meteorites [e.g., 1-6], has been used to constrain the efficiency of mixing in the protosolar nebula. To understand these large scale processes, a better knowledge is needed of the fine-scale isotopic heterogeneity existing within individual unmetamorphosed meteorites. Sequential leaching experiments, in which acids of differing composition and strength are used to successively dissolve different phases, have permitted the identification of fine-scale nucleosynthetic anomalies of Cr [7], Ba [8, 9] Zr [10] and Mo [11]. We report here preliminary results of sequential leaching of the Murchison carbonaceous chondrite. This meteorite was chosen for study both because of its primitive, unequilibrated nature (class CM2) and because a previous study [8] has demonstrated that it contains large internal nucleosynthetic anomalies in Ba.

Analytical techniques: About 16.5 grams of powdered Murchison meteorite were subjected at the University of Chicago to a 6-step leaching procedure:
1) 50 ml acetic acid + 50 ml H_2O, 1 day, 20^oC
2) 25 ml HNO_3 + 50 ml H_2O, 5 days, 20^oC
3) 30 ml HCl + 35 ml H_2O, 1 day, 75^oC
4) 30 ml HF + 15 ml HCl + 15 ml H_2O, 1 day, 75^oC
5) 10 ml HF + 10 ml HCl, 3 days, 150^oC
6) 2 ml HNO_3 15M + 2 ml HF 28M, 120^oC, 1 night
(This last step was intended to dissolve the residue, but probably did not dissolve highly refractory phases). A fraction of each leachate was taken for Os isotopic analysis. About 10% of each fraction was spiked with ^{190}Os, ^{185}Re and ^{196}Pt for concentration determinations; the rest was used for high precision Os isotopic measurements. About 90% of step 1 was unintentionally spiked, while only 10% was unspiked. Both fractions were analyzed in high precision runs, and a correction was made for the very small amount of spike present in the 90% aliquot. After drying down if needed to remove HF and acetic acid, all fractions except #6 were sealed in Carius tubes in HNO_3:HCl (2:1), and heated at 230^oC for 24 hours [12]. Fraction 6 was oxidized by adding Cr^{VI} in H_2O and heating at 90^oC overnight.

Os was extracted with liquid bromine and purified by microdistillation [13]. Os isotopic compositions for concentration calculations were determined by NTIMS [14,15] on the CRPG Finnigan MAT262. High precision Os isotopic compositions were determined by NTIMS from the unspiked aliquots (and the large spiked aliquot of #1) using the University of Durham Triton. In-run oxygen compositions were used for oxide corrections. Potential interferences from PtO_2, WO_3, and ReO_3 were monitored closely on the SEM before and after all runs, and any necessary corrections applied. In nearly all cases, the only interferences were from PtO_2. Mass bias corrections assumed a $^{192}Os/^{189}Os$ ratio of 2.5277. Results are reported as ε values, where $\varepsilon_i=(10^4*(^iOs/^{189}Os_{sample} - {}^iOs/^{189}Os_{std})/^iOs/^{189}Os_{std})$. Reproducibilites (2σ) in ε units of the Durham standard were 0.64 for ε_{186}, 0.09 for ε_{188}, and 0.06 for ε_{190}.

Results: Fig. 1 shows the distribution of Os among the leachate fractions. The summation of the Os in each fraction indicates a total Os concentration of about 550 ppb, which given the expected heterogeneity, is similar to previous determinations (759 ppb [16]; 607 ppb [17]) of the Murchison Os content.
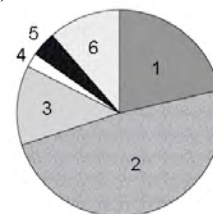

Figure 1. Osmium distribution among the leachate fractions.

Os isotopic anomalies relative to the Durham terrestrial standard are shown in Fig. 2a. The observed spectra closely match those expected for deficiencies (negative anomalies, fractions 1 and 5) or excesses (positive anomalies, fractions 2, 3 and 4) of a component enriched in Os derived from the s-process. Spectra 4 and 5 were obtained from very low intensity runs of small quantities of Os and should be interpreted with caution until reanalysis. Nevertheless, even these low intensity runs show spectra consistent with the over or underabundance of an s-rich component. These spectra are also consistent with those obtained from bulk analyses of the Tagish Lake chondrite [5], which show a deficiency in the s-component.
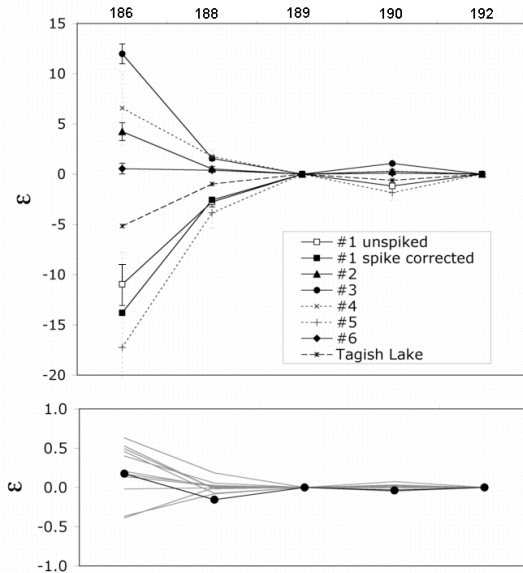
Figure 2. a) Os isotopic compositions of the leachates relative to the Durham terrestrial standard. ε_{186} values were not corrected to initial solar system values since the time integrated Pt/Os ratio of the standard is unknown, but this should not change the ε_{186} by more than 0.1 unit (see on-line material in [5]). Tagish Lake data from [5]. b) Weighted average of the leachate fractions compared to standard analyses.

Weighted averages of ε_{186}, ε_{188}, and ε_{190} of the fractions are all less than 0.2 (Fig. 2b), despite the extreme heterogeneity of the individual leachates. Thus the bulk Os composition of Murchison is very close to that of most chondrites and of earth. Among the leachates, ε_{186}, ε_{188}, and ε_{190} correlate well (Fig. 3), consistent with addition or subtraction of an *s*-process component. Slight deviations from the correlations likely reflect difficulties due to analysis of small quantities of Os. Repeat analyses of larger quantities are underway.

Discussion: These results provide evidence for extreme isotopic heterogeneity within Murchison, consistent with inhomogeneous distribution of a component rich in *s*-process Os. Nevertheless, the overall Os composition obtained from the weighted average of the fractions is very similar to that of most chondrites and earth. Interestingly, the *s*-process rich Os component is accessed by relatively mild leaches (fractions 2, 3 and 4). This suggests that it may not be mainly carried by presolar SiC grains, as suggested for the Tagish Lake meteorite [5], but rather by more easily digestible presolar phases such as graphite or metal alloy.

Analyses of larger quantities of Os, currently underway, should allow us to more precisely define the isotopic composition of the *s*-process component. This will place constraints on the temperature and the neutron density of the stellar environment in which this component formed.
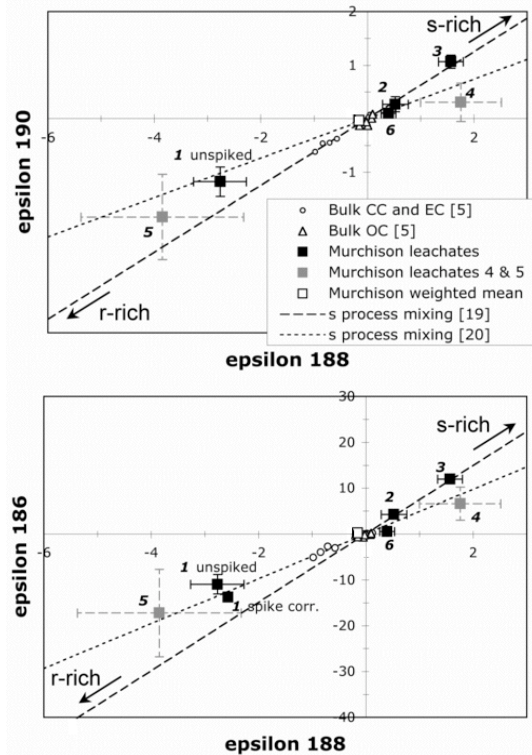

Figure 3. Correlations between a) ε_{190} and ε_{188} and b) ε_{186} and ε_{188} among the various leachate phases. Results from bulk analyses [5] of carbonaceous (CC), enstatite (EC) and ordinary (OC) chondrites are shown for comparison. Dashed and dotted lines show the effect of addition or subtraction of an *s*-process component, calculated using a formula analogous to Eq. 1 of [18] and assuming two different models for the *s*-process Os isotopic composition [19, 20].

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