UNDERSTANDING VARIOUS CONTRIBUTIONS TO THE CHROMIUM ISOTOPIC COMPOSITION OF METEORITES, AND THEIR IMPLICATIONS FOR Mn-Cr CHRONOLOGY.

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Introduction: Previous studies have shown that ^{54}Cr is heterogeneously distributed in Solar System materials, including chondrites, differentiated stony and iron meteorites [1]. Each meteorite group has its characteristic ^{54}Cr signature. Even though the ^{54}Cr anomalies are widely thought to be nucleosynthetic in origin, the nucleosynthetic process responsible for its production and the cause of the heterogeneous distribution of these nucleosynthetic products remain mysterious. No correlation with other iron-group elements has yet been found, except possibly with ^{46}Ti and ^{50}Ti [2]. The carrier phases of the ^{54}Cr anomalies have never been isolated or identified. Even though CAIs in carbonaceous chondrites (CC) have high ^{54}Cr [3,4], the low abundance of CAIs in CCs and their low Cr concentration means that they cannot account for the anomaly observed in the bulk meteorites. Step-wise acid dissolution experiments have revealed that there are both ^{54}Cr-poor and ^{54}Cr-rich carrier phases [5,6]. The former is soluble in relatively weak acid and the latter is more acid-resistant. In our recent work, we attempted to concentrate ^{54}Cr-rich phases in insoluble organic matter (IOM) rich residues through CsF digestion of the sample, the same technique used to concentrate pre-solar grains. Large positive ^{54}Cr anomalies up to 200 ε were found in the hot HCl leachates of the IOM of relatively primitive CCs [7]. IOM of primitive ordinary chondrites (OC) also show positive ^{54}Cr anomalies up to 40 ε in their leachates, in contrast to the small negative ^{54}Cr anomalies (~ -0.4 ε) in the bulk rocks.

Another concern is whether the existence of ^{54}Cr anomalies compromise the use of the short-lived chronometer ^{53}Mn-^{53}Cr. Correlated excesses in ^{53}Cr and ^{54}Cr have been found for CCs [8]. The HCl-leachates of IOMs of chondrites, however, show very small negative ^{53}Cr anomalies of 2 ε or less in spite of large ^{54}Cr excesses [7] suggesting that the ^{54}Cr anomalies do not significantly affect ^{53}Cr abundances. The Cr isotopic compositions of some meteorites, or fractions thereof, also may have been affected by spallation of target elements (e.g., Fe) during their exposure to galactic cosmic rays. Spallation can produce both ^{53}Cr and ^{54}Cr, and further complicate the Cr systematics in meteorites.

In this study, we have reexamined the Cr isotope systematics in various types of bulk meteorites. We also took a closer look at the IOM in chondrites to try to better characterize the nucleosynthetic effect. A series of meteoritic metal samples including pieces of iron meteorites and metals separated from chondrites and one pallasite, were studied. These metal samples have high Fe/Cr and make them ideal for studying cosmogenic effects.

Samples and Methods: Powdered bulk stony meteorite samples were fused with lithium borates at 1050~1100 °C and subsequently dissolved in 2 N HNO_3. The metal samples were dissolved in a mixture of HCl and HNO_3 (in 2:1 ratio by volume). Aliquants of IOM had been processed previously for Os isotope study [9]. AR4 was combusted in a Carius tube at 1000 °C. Another aliquant of the same residue was leached with 6 N HCl at 80 °C. The leachate is designated as "HHLL". Its residue was combusted in the same way as above and designated as "HHLR".

Chemical separation of the Cr followed the method described in [7], except for iron meteorites and one pallasite metal. Because of the extremely high Fe/Cr ratios in these samples, they were passed through an anion-exchange column to remove Fe before the cation exchange procedure [7]. The Cr isotopes were analyzed with a Triton TIMS at DTM [7].

Results and Discussions: *Bulk meteorites and terrestrial samples*. The external reproducibility for ε^{53}Cr and ε^{54}Cr over a period of 8 months for NIST SRM 3112a Cr is 0.08 and 0.13 ε. We find a difference of 0.25 ε for ε^{54}Cr between this standard and our previous Cr standard, which we attribute to chemically-induced mass fractionation of the Cr during purification. As shown in Fig. 1, most terrestrial rocks show identical Cr isotopic compositions to NIST 3112a. The Cr isotopic data for chondrites are generally consistent with recent studies [1,10]: All chondrites show higher ε^{53}Cr values than the standard; Most enstatite chondrites (EC) show no ^{54}Cr anomaly relative to NIST 3112a, CCs have positive anomalies up to 1.6 ε, and OCs show slightly negative anomalies of ~ -0.4 ε. ^{54}Cr excesses in CCs are grossly correlated with that of ^{53}Cr and with measures of volatility, such as matrix abundance and Zn/Si ratio, indicating these Cr excesses are controlled by volatility of the samples. One E-chondrite, QUE94204, has resolvably higher ε^{53}Cr and ε^{54}Cr than the rest of the group. We will show later that this can be at least partially explained by cosmogenic effects.

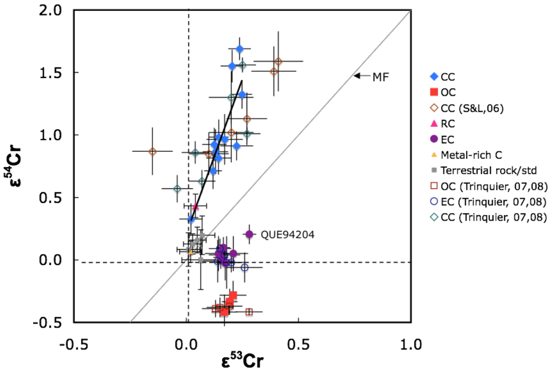

Fig. 1. Cr isotopic data of chondrites and terrestrial rocks.

IOM of CCs. Both the leachate and residue of Allende IOM show only small ^{54}Cr anomalies, comparable with the bulk meteorite (0.9 ε) (Table 1). This supports our conclusion [7] that the anomalous Cr carrier phases in more highly metamorphosed chondrites were mostly destroyed. The acid leachate (HHLL) of Murchison IOM shows a large positive ^{54}Cr anomaly of 136 ε, and a small ^{53}Cr deficit. The carrier phases in the residue (HHLR) have a similar Cr isotopic composition to the leachate, except that the anomalies are diluted by "normal" Cr. The bulk Murchison IOM has a ^{54}Cr anomaly of 20 ε, representing 6% of the total Cr in the rock. Thus we were able to concentrate at least one of the ^{54}Cr carrier phases in IOM. SEM imaging revealed that the mineral phases in IOM are dominated by chromites and Mg-rich spinels.

Table 1. Cr isotopic compositions of IOM.

	ε^{53}Cr	ε^{54}Cr	Cr (%)
Allende HHLL	0.08±0.11	0.42±0.26	0.7
Allende HHLR	-0.10±0.07	1.50±0.13	3.4
Allende AR4	0.00±0.05	1.57±0.12	4.1
Murchison HHLL	-1.88±0.06	136.56±0.14	0.2
Murchison HHLR	-0.48±0.07	20.49±0.17	5.8
Murchison AR4	-0.55±0.08	21.85±0.18	6.0

Fractions of OCs. In a search for ^{54}Cr-poor carrier phases, different fractions of Tieschitz (H3), including matrix, two types of chondrules (magnetic and non-magnetic) and metal grains (in the matrix) were analyzed. All fractions have similar Cr isotopic compositions as the bulk meteorite, except the metal. The metal exhibits positive ε^{53}Cr and ε^{54}Cr values of 0.44 and 0.79 respectively. The anomalies were partially caused by cosmogenic effects, which will be discussed in the next section.

Metals. Among the metals of chondrites analyzed, QUE94204 shows the highest ^{53}Cr and ^{54}Cr anomalies of 3.89 ε and 14.08 ε. Metal from the pallasite Brenham shows large ^{53}Cr and ^{54}Cr anomalies of 19.4 ε and 73.56 ε. Three pieces of Carbo, sampled from different locations within the same bar, show a strong correlation between ε^{54}Cr and ε^{53}Cr (Fig. 2). Brenham has an exposure age of 200 Ma, and that of Carbo is 850 Ma. The slope in Fig. 2 is 4, consistent with production rates of the two isotopes by cosmogenic effects [11]. The Cr isotope anomalies of Carbo vary by almost a factor of two within a distance of 15cm, reflecting shielding effects. Even though most stony meteorites have low exposure ages, these results indicate that Cr in phases with high Fe/Cr may show cosmogenic effects. The higher ε^{53}Cr and ε^{54}Cr values of the WR analysis of QUE94204 also may be partially explained by cosmogenic effects (Fig. 1). These results also suggest caution in the use of high Fe/Cr minerals to construct the Mn-Cr isochrons in some meteorites and the need for precise measurement of ^{54}Cr/^{52}Cr in these phases.

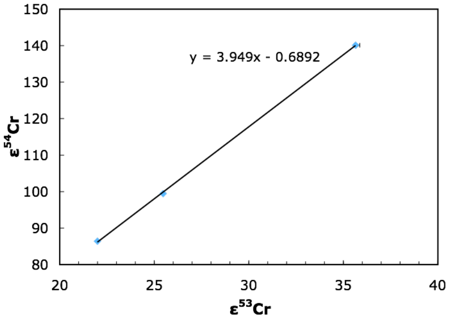

Fig. 2. Cr isotopic compositions of pieces of Carbo from different depths beneath the meteorite surface.

Conclusions: ^{54}Cr variations have been confirmed for bulk chondrites. The ^{54}Cr carrier phases in leachate and residue of Murchison IOM may share similar Cr isotopic compositions. Cosmogenic effects were documented for samples from an iron meteorite. These variations in Cr isotope compositions suggest caution in the use of Mn-Cr chronometer, although the nucleosynthetic ^{53}Cr variation is likely to be very small.

References: [1] Trinquier A. et al. (2007) *ApJ, 655*, 1179-1185. [2] Trinquier A. et al. (2008) *GCA*, 72, A956. [3] Birck J.-L. and Allègre J. (1984) *GRL*, *11*, 943-946. [4] Papanastassiou D. A. *ApJ, 308*, L27-L30. [5] Rotaru M. et al. (1992) *Nature, 358*, 465-470. [6] Podosek F. A. et al. (1997) *MAPS, 32*, 617-627. [7] Qin L. et al. (2008) *LPS XXXIX*, Abstract #2078. [8] Shukolyukov A. and Lugmair G. W. (2006) *EPSL, 250*, 200-213. [9] Yokoyama T. et al. (2008) *LPS XXXIX*, Abstract #1376. [10] Trinquier A. et al. (2008) *GCA, 72*, 5146-5163. [11] Birck J.-L. and Allègre C. J. (1985) In *Isotopic Ratios in the Solar System*, pp. 21-25. Cepadues-Editions.