Nucleosynthetic strontium isotope anomalies in carbonaceous chondrites

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A B S T R A C T

Precise Sr isotopic compositions in samples from sequential acid leaching experiments have been determined for three carbonaceous chondrites, Allende, Murchison, and Tagish Lake, together with those in the bulk aliquots of these meteorites. The chondritic acid leachates and residues were characterized by Sr isotope anomalies with variable $\mu^{84}Sr$ values (10^6 relative deviation from a standard material) ranging from $+120$ to -4700 ppm, documenting multiple nucleosynthetic sources within a single meteorite. In addition, the $\mu^{84}Sr$ patterns across leaching samples for individual chondrites differed from one another. The highest $\mu^{84}Sr$ values were observed for leaching Step 3 (HCl+H_2O, 75 °C) for Allende and Murchison likely because of the incorporation of calcium and aluminum-rich inclusions (CAIs). In contrast, extremely low $\mu^{84}Sr$ values were observed in the later fractions (Steps 6 and 7) for Murchison and Tagish Lake, suggesting the existence of s-process-enriched presolar SiC grains derived from AGB stars.

A $\mu^{84}Sr$–$\varepsilon^{54}Cr$ diagram was prepared with the CAIs and bulk aliquots of carbonaceous chondrites and other meteorites (noncarbonaceous) that were plotted separately; however, they still formed a global positive correlation. CAIs presented the highest $\mu^{84}Sr$ and $\varepsilon^{54}Cr$ values, whereas carbonaceous chondrites and noncarbonaceous meteorites had intermediate and the lowest $\mu^{84}Sr$ and $\varepsilon^{54}Cr$ values, respectively. The positive trend was interpreted as resulting from global thermal processing in which sublimation of high $\mu^{84}Sr$ and $\varepsilon^{54}Cr$ carriers generated the excess $\mu^{84}Sr$ and $\varepsilon^{54}Cr$ signatures in CAIs, while noncarbonaceous planetesimals accreted from materials that underwent significant thermal processing and thus had relatively low $\mu^{84}Sr$ and $\varepsilon^{54}Cr$ values. Apart from the global trend, the carbonaceous chondrites and noncarbonaceous meteorites both exhibited intrinsic variations that highlight an isotopic dichotomy similar to that observed in other isotope combinations (e.g., $\varepsilon^{54}Cr$–$\varepsilon^{50}Ti$, $\varepsilon^{54}Cr$–$\Delta^{17}O$). A plausible scenario for creation of the intrinsic variations involves local thermal processing (e.g., flash heating for chondrule formation) caused by additional selective destruction of presolar grains differently than that caused by global thermal processing. The existence of such a global positive trend and local variations for two meteorite groups suggests a complicated dynamic history for the dust grains with respect to thermal processing, material transportation, and mixing in the protoplanetary disk prior to planetesimal formation.

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1. Introduction

An increasing amount of highly precise stable isotope data for heavy elements is becoming available for a variety of chondrites and differentiated meteorites. These data point to the existence of nucleosynthetic isotope heterogeneity in the protoplanetary disk (e.g., Ti, Cr, Ni, Mo, Ru, Ba, Nd, and Sm: Andreasen and Sharma, 2006; Burkhardt et al., 2011; Carlson et al., 2007; Chen et al., 2010; Dauphas et al., 2002a; Qin et al., 2010; Regelous et al., 2008;

Trinquier et al., 2007, 2009), yet there are conflicting results that support homogeneous isotope distribution in the early solar system (e.g., Cd, Te, Hf, and Os: Fehr et al., 2005; Sprung et al., 2010; Wombacher et al., 2008; Yokoyama et al., 2010, 2007). This inconsistency may arise as a consequence of incomplete mixing of dust grains and/or destruction of some selected presolar materials during thermal processing in the solar nebula (Dauphas et al., 2010; Trinquier et al., 2009). However, the details regarding the processes that may have led to isotope heterogeneity are not totally understood. A key issue is to identify the main presolar phases and nucleosynthetic origins for the individual elements that contributed to the solar system. Sequential acid leaching of powdered meteorites is a useful approach for detecting internal isotope

anomalies in meteorites, particularly for trace elements with concentrations of a few ppm in the presolar grains. Previous acid leaching studies of bulk chondrites for refractory heavy elements revealed the existence of multiple presolar phases from diverse nucleosynthetic sources within single meteorites (e.g., Cr, Zr, Mo, Sr, Ba, Nd, Sm, W, and Os: Burkhardt et al., 2012a; Dauphas et al., 2002b; Hidaka et al., 2003; Paton et al., 2013; Qin et al., 2011a; Reisberg et al., 2009; Rotaru et al., 1992; Schönbächler et al., 2005; Trinquier et al., 2007).

In this study, stable Sr isotopic anomalies in three carbonaceous chondrites were investigated using the sequential acid leaching technique. Strontium has four isotopes produced by the stellar nucleosynthesis s-process (^{86}Sr, ^{87}Sr, and ^{88}Sr), r-process (^{87}Sr and ^{88}Sr), and p-process (^{84}Sr). In addition, radioactive decay of ^{87}Rb ($T_{1/2} = 4.88 \times 10^{10}$ yr) contributes to the abundance of ^{87}Sr. Stable Sr isotope anomalies in meteorites were first found in calcium and aluminum-rich inclusions (CAIs) from the Allende meteorite using thermal ionization mass spectrometry (TIMS) (Papanastassiou and Wasserburg, 1978). The anomalous inclusions, which had extremely low ^{84}Sr/^{86}Sr ratios (-0.4‰ relative to a terrestrial standard), are now known as the "FUN inclusions" (fractionation and unknown nuclear inclusions). Patchett (1980b) also analyzed Sr isotope compositions in chondrules and CAIs from Allende using the double-spike technique with TIMS, and discovered that Sr isotopic variations observed in the chondrite components were solely the result of mass-dependent fractionation that occurred in the solar system. Strontium isotope compositions in presolar SiC fractions separated from the Murchison meteorite (CM2) were measured by Podosek et al. (2004). They concluded that the SiC grains were enriched in Sr isotopes produced by the s-process in AGB stars.

More recently, Sr isotope compositions in bulk meteorites and their components (e.g., CAIs, acid leachates) were precisely determined using new generation TIMS instruments (Andreasen and Sharma, 2007; Hans et al., 2013; Moynier et al., 2012; Paton et al., 2013). Moynier et al. (2012) reported that carbonaceous chondrites had variable ^{84}Sr/^{86}Sr ratios that were at a 0.5–1ε level higher than ordinary and enstatite chondrites as well as samples from Earth, Mars, the Moon, and HED meteorites. More importantly, Allende CAIs were characterized by the highest ^{84}Sr/^{86}Sr ratios ($>1\varepsilon$ higher than the terrestrial sample). Similar results were obtained by Paton et al. (2013). Such Sr isotopic variations among diverse planetary bodies suggest incomplete mixing of Sr isotopes in the early solar nebula.

In the present study, to further identify the isotopically anomalous carriers for Sr in chondrites, sequential acid leaching experiments were performed for bulk rocks of three different types of carbonaceous chondrites (Allende, Murchison, and Tagish Lake (C2 ungrouped)), and the precise Sr isotope compositions in the leachates and residues were determined using the state-of-the-art TIMS analysis technique. The results were anticipated to provide key information regarding the origin of Sr isotope anomalies in chondrites necessary for clarifying the origin of the planetary scale Sr isotope diversity observed in meteorites and their components.

2. Experimental

Details of the sample description, chemical procedures, and mass spectrometry for Sr isotope analysis are given in the supplemental on-line material (SOM). Only the essence of the experiments is briefly described below.

Aliquots (1.0–1.6 g) of powdered Allende (Allende-A, Allende-B, and Allende-C), Murchison, and Tagish Lake (Tagish Lake-A and Tagish Lake-B) were subjected to a six-step sequential acid leaching following the method described in Reisberg et al. (2009).

Step 1) 30% $CH_3COOH + H_2O$, 20 °C

Step 2) 16 M $HNO_3 + H_2O$, 20 °C
Step 3) 12 M $HCl + H_2O$, 75 °C
Step 4) 30 M $HF + 12$ M $HCl + H_2O$, 75 °C
Step 5) 30 M $HF + 12$ M HCl, 150 °C
Step 6) 16 M $HNO_3 + 30$ M HF, 120 °C.

For the Allende-C, Murchison, and Tagish Lake-B powders, the final residues remaining after Step 6 were combusted in quartz Carius tubes at 1000 °C, followed by acid digestion using $HF + HNO_3$ in a high-pressure Digestec DAB-2 (Berghof) sample digestion system (referred to as Step 7). For the Allende-C, Murchison, and Tagish Lake-B samples, an aliquot ($\sim$5%) of each leaching fraction (Steps 1–7) was removed prior to the Sr purification process to determine the abundance of 22 lithophile trace elements (Rb, Sr, Y, Cs, Ba, REEs, Pb, Th, and U) using a quadrupole-type inductively coupled plasma mass spectrometer (X-Series II, ThermoFisher Scientific).

Bulk aliquots of Allende, Murchison, and Tagish Lake (200–600 mg) were digested for Sr isotope analysis using a newly developed, aggressive acid decomposition procedure with $HF + HNO_3 + H_2SO_4$ in the DAB-2 operated at 250 °C. This procedure confirms complete digestion of acid-resistant carbonaceous presolar grains (e.g., SiC). To measure the 22 trace elements in bulk meteorites, additional aliquots of these meteorites (50 mg each) were separately decomposed via conventional acid digestion using $HF + HNO_3 + HClO_4$ (Yokoyama et al., 1999).

High-precision Sr isotope measurements were performed using TIMS (Triton-*plus*, ThermoFisher Scientific). The purified Sr (10–500 ng) was loaded onto a single outgassed W filament using a Ta_2O_5 activator slurry. The results were obtained by averaging 100–400 ratios (2σ rejection level = 4.55% of the data) obtained in the static multicollection mode using five Faraday cups. The analytical error within a single run was evaluated using 2SE.

The Sr isotope ratios were normalized to ^{86}Sr and corrected for mass fractionation by assuming ^{86}Sr/^{88}Sr = 0.1194. Because of the long-term fluctuations in the ^{84}Sr/^{86}Sr and ^{87}Sr/^{86}Sr ratios observed during the course of this study, the entire analytical period was divided into 9 analytical campaigns (see SOM). The analytical uncertainties (2SD) of the Sr isotope ratios in the individual analytical campaigns ranged from 20 to 34 ppm for ^{84}Sr/^{86}Sr and 1 to 8 ppm for ^{87}Sr/^{86}Sr (Fig. S1). The problem of long-term fluctuations was reconciled by normalizing each observed ^{84}Sr/^{86}Sr ratio to the average of the ^{84}Sr/^{86}Sr ratios for the NIST 987 standard determined in the same analytical campaign as follows:

$$\mu^{84}Sr = \left[\frac{(^{84}Sr/^{86}Sr)_{sample}}{(^{84}Sr/^{86}Sr)_{NIST987}} - 1 \right] \times 10^6. \tag{1}$$

Likewise, the ^{87}Sr/^{86}Sr ratio for each sample was recalculated by normalizing it to the generally accepted ^{87}Sr/^{86}Sr value of 0.71025 for NIST 987 as follows:

$$^{87}Sr/^{86}Sr = \frac{(^{87}Sr/^{86}Sr)_{sample}}{(^{87}Sr/^{86}Sr)_{NIST987}} \times 0.71025, \tag{2}$$

where $(^{87}$Sr$/^{86}$Sr$)_{NIST987}$ is the average of the ^{87}Sr/^{86}Sr ratios for NIST 987 determined in the same analytical campaign. The validity of this approach was confirmed by the consistency of the μ^{84}Sr values in a terrestrial basalt (JB-3) analyzed for the different analytical periods (Table 1).

3. Results

3.1. Strontium abundance in the acid leachates and residues

The absolute and relative abundances of 22 trace elements in the individual leaching fractions and the final residues are listed in