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Sr stable isotope composition of Earth, the Moon, Mars, Vesta and meteorites

Frédéric Moynier [a,b,*], Arnaud Agranier [c], Dominik C. Hezel [d], Audrey Bouvier [e]

[a] Department of Earth and Planetary Sciences, Washington University in St Louis, One Brookings Drive, St Louis, MO 63130, USA
[b] McDonnell Center for Space Sciences, Washington University in St Louis, One Brookings Drive, St Louis, MO 63130, USA
[c] Institut Universitaire Européen de la Mer, Université de Bretagne Occidentale, Domaines Océaniques, UMR 6538, Place Nicolas Copernic 29820 Plouzané, France
[d] IARC, Department of Mineralogy, Natural History Museum, Cromwell Road, London, SW7 5BD, UK
[e] Arizona State University, School of Earth and Space Exploration, Tempe, AZ 85287-1404 USA

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ABSTRACT

High-precision stable Sr isotopic variations (^{88}Sr/^{86}Sr) are reported in a variety of terrestrial samples, martian and lunar meteorites, HED, undifferentiated primitive meteorites, chondrules and refractory inclusions. Almost all the whole-rock samples are isotopically indistinguishable at a 50 parts per million (ppm) level. The exceptions are CV and CO chondrites which are isotopically light and for which we believe that their isotopic composition is controlled by the proportion of refractory material. Five separated chondrules and one refractory inclusion from Allende are isotopically light, with $\delta^{88/86}$Sr fractionations up to −1.73‰, whereas the matrix is enriched in the heavy isotopes ($\delta^{88/86}\mathrm{Sr} = +0.66$‰). The depletion in heavy isotopes observed in chondrules and refractory inclusions could be attributed to the condensation of a material already depleted in Sr, however, in that case more than 60% of the original material would be unaccounted. We propose instead that isotopic fractionation by electromagnetic sorting of ionized heavy Sr from neutral Sr in the early solar system for the origin of the fractionation observed in refractory inclusions and redistribution of Sr by aqueous alteration for the origin of the fractionation observed in chondrules and matrix. We conclude that CV and CO chondrites are not the primary building blocks for Earth and Mars.

1. Introduction

The recent development of multiple-collector inductively-coupled plasma mass-spectrometry (MC-ICP-MS) has allowed the measurement of small isotopic fractionation for “heavy” elements during high temperature processes. Notably Mg (Jacobsen et al., 2008; Norman et al., 2004; Pearson et al., 2006; Teng et al., 2010; Wiechert and Halliday, 2007; Young and Galy, 2004), Fe (Dauphas et al., 2009; Hezel et al., 2010; Mullane et al., 2005; Needham et al., 2009; Poitrasson et al., 2004; Schoenberg and Blanckenburg, 2006; Shahar et al., 2008; Weyer et al., 2005; Williams et al., 2009; Zhu et al., 2001), Cu (Herzog et al., 2009; Luck et al., 2003; Moynier et al., 2006a; 2010a), Zn (Herzog et al., 2009; Luck et al., 2005; Moynier et al., 2006a; 2009a), Ni (Cook et al., 2007; Moynier et al., 2007) and Ca (Russel et al., 1978; Simon and DePaolo, 2010) are providing a new perspective on metal/silicate/sulfide/vapor separation during nebular and planetary processes. With the exception of oxygen isotopes which are clearly heterogeneous in the solar system (Clayton, 1993), and also Cr (Podosek et al., 1997; Qin et al., 2010; Rotaru et al., 1992; Shukolyukov and Lugmair, 2006; Trinquier et al., 2007) and Ti (Leya et al., 2008; Trinquier et al., 2009), the initial inventories of most of the other elements seem to be derived from homogeneous reservoirs (Fehr et al., 2006; Luck et al., 2005; Molini-Velsko et al., 1986; Moynier et al., 2009b; Simon and DePaolo, 2010; Wombacher et al., 2008; Zhu et al., 2001) and their isotopic variations are due to mass-dependent fractionations controlled by phase separation processes. Recent improvements in analytical methods and instrumentation permit resolution of small (<20 ppm) isotopic anomalies (non mass-dependent isotopic fractionation) for several elements (e.g. Ca, Ni, Mo, Ru, Ba, Sm, Nd, and W) in bulk meteorites (Andreasen and Sharma, 2006; Andreasen and Sharma, 2007; Carlson et al., 2007; Chen et al., 2010; Dauphas et al., 2002; Qin et al., 2008; Ranen and Jacobsen, 2006; Simon et al., 2009; Yin et al., 2002).

Strontium is a refractory lithophile element with a 50% condensation temperature (Tc) of 1464 K (Lodders, 2003) and has 4 stable isotopes, ^{84}Sr (0.56%), ^{86}Sr (9.86%), ^{87}Sr (7.00%) and ^{88}Sr (82.58%). Part of ^{87}Sr is the product of the long-lived radioactive decay of ^{87}Rb, and measuring ^{87}Sr/^{86}Sr ratio together with Rb/Sr ratio has been one of the most successful chronometers and tracers in Earth Sciences. For Rb–Sr chronology the ^{88}Sr/^{86}Sr ratio is usually used as a normalization ratio to correct for instrumental mass-dependent effect on thermo-ionization mass-spectrometry (TIMS). The preferential interest in radiogenic ^{87}Sr and the difficulty using TIMS to get precise mass-dependent variations left the fractionation patterns of the stable Sr isotopes essentially untouched. By employing the double spike technique combined with TIMS, Patchett (1980a,1980b) was able to

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* Corresponding author. Department of Earth and Planetary Sciences, Washington University in St Louis, One Brookings Drive, St Louis, MO 63130, USA.
E-mail address: moynier@levee.wustl.edu (F. Moynier).

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correct for instrumental mass fractionation and found that chondrules and refractory inclusions of the Allende meteorites have a $^{88}Sr/^{86}Sr$ ratio depleted up to 2.8‰ in light isotopes of Sr when compared to the bulk meteorites and terrestrial samples. These enrichments in light isotopes are puzzling since most refractory inclusions demonstrate clear evaporation loss and recondensation as recorded by Si and Mg isotopic compositions (Richter et al., 2002; Shahar and Young, 2007); and the mass-dependent isotopic fractionation theory predicts that condensed phase should be enriched in heavy isotopes. That kind of enrichment in the heavy isotopes in the refractory phase has notably been observed for O, Mg, Si, K, Ca, Ti, Fe and Zn during experimental evaporation of material with solar compositions (Davis et al., 1995; Esat et al., 1986; Floss et al., 1996; Molini-Velsko et al., 1986; Uyeda et al., 1991; Wang et al., 2001; Xue et al., 1996). In addition to Sr, few other elements show enrichments in light isotopes in refractory inclusions, notably Ca (Niederer and Papanastassiou, 1984), Eu (Moynier et al., 2006b), Zn (Luck et al., 2005; Moynier et al., 2007), and Cd (Wombacher et al., 2008). Possible mechanisms at the origin of these enrichments in light isotopes may involve kinetic effect (Richter, 2004; Simon and DePaolo, 2010) or electromagnetic separation (Moynier et al., 2006b).

Here we investigate the degree of stable isotopic fractionation of Sr in a variety of terrestrial samples, martian and lunar meteorites, HED meteorites (supposedly coming from the asteroid 4-Vesta) and chondrites (ordinary, carbonaceous and enstatite) as well as in separated chondrules and one calcium aluminum-rich inclusion (CAI) from the Allende CV3 carbonaceous chondrite. The Sr isotope compositions were measured by MC-ICP-MS which has been proved to be the best instrument to analyze the stable isotope composition of Sr (de Souza et al., 2010; Fietzke and Eisenhauer, 2006; Fortunato et al., 2004; Ohno and Hirata, 2007).

2. Samples and analytical methods

2.1. Sample description

Since little is known on the stable Sr isotope composition of the Earth, a variety of geological reference materials from different magmatic settings representing the main reservoirs of Sr on Earth were analyzed, namely USGS BHVO-2, an ocean island basalt from Hawaii, USA; USGS AGV-1, an andesite from the Guano Valley, in Lake County, Oregon, USA, BCR-2, a basalt from the Columbia River, 29 miles east of Portland, Oregon, USA. The USGS granite G2 and a S-type graniodorite S2 from Cowra, Australia (Chappell et al., 1993) were also analyzed to further test the effect of magmatic differentiation on the Sr isotope composition.

The meteorite samples were selected to cover a wide range of planetary bodies: three Martian shergottites — Zagami, Elephant Moraine A79001 (EETA 79001) and Sayh al Uhaymir 008 six carbonaceous chondrites from different groups — Orgueil (CI), Tagish Lake (C2-ungr), Ornans (CO), Murchison (CM), Allende and Mokoia (CV); four ordinary chondrites — Forest Vale and St Marguerite (H4), Forest City (H5) and Tuxtuac (LL5); four enstatite chondrite — Indarch (EH4), Abee (EH4), Qingzhen (EH3) and Atlanta (EL6), one aubrite — Norton County; two HEDs — Palo Blanco Creek (eucrite) and Aioun el Atrouss (diogenite); one lunar anorthositic breccia meteorite — MacAlpine Hills 88105 (MAC 88105).

In addition we analyzed 5 separated chondrules, a fragment of one single CAI, and one matrix aliquot from Allende (CV). The chondrules were separated by gently crushing the meteorite using a hammer. Chondrules were then handpicked under a binocular microscope. SEM studies on chondrules not used for this study show that these are free of any matrix material (e.g. Hezel et al., 2010). A fine-grained CAI was extracted from a slab of the Allende meteorite using stainless steel tools at the Center for Meteorite Studies at ASU. The composition in trace elements measured on a ~50 mg bulk fragment (unleached) of this same inclusion reveals a pattern of chemical group II REE (Brearley and Jones, 1998).

2.2. Analytical methods

For every bulk sample, 100–200 mg of homogenous powder was dissolved in a 4:1 HF/HNO_3 mixture. Individual chondrules weighted from 1 mg to and 3 mg and were directly dissolved in HF/HNO_3 with no preliminary grinding. The chondritic samples were dissolved under pressure in Parr bombs in an oven at 130 °C whereas the terrestrial samples, individual chondrules as well as the achondrites were dissolved in closed Teflon beakers at 130 °C for several days. After evaporation of the HF/HNO3, the samples were re-dissolved in aqua regia at 130 °C for several days in order to ensure the total dissolution of fluorides.

In all cases, the residue was re-dissolved in 4 N HNO_3 and was loaded on Teflon micro-columns with 300 µl of Sr specific resin (Eichrom) following the procedure described in Fietzke and Eisenhauer (2006). The column was further washed by 4 ml of 4 N HNO_3. The Sr was finally eluted in 3 ml of warm (40–50 °C) H_2O. This procedure was repeated twice to ensure a clean Sr fraction. Extraction yield were $>98\%$ and Sr blanks <0.1 ng, which is usually negligible with respect to the amount of Sr processed from the samples (~1 µg).

A fragment of ~50 mg of a fine-grained CAI extracted from the Allende chondrite was leached in cold 1 M HF (10 mn), 2.5 M HCl (10 mn) in ultrasonic bath, and rinsed in Milli-Q H_2O which is sufficiently mild to remove the surface contamination and not affect the chemical composition of the insoluble minerals (e.g., silicates, oxides). The residue (~45 mg) was fully dissolved using a Parr bomb and the fraction containing Sr was collected in 2.5 M HCl during the AG50W-X8 cation resin step following the protocol described in Bouvier et al. (2008) before further Sr purification as described above.

The samples were analyzed for their Sr isotopic compositions by MC-ICP-MS, on a Thermo Fisher Neptune at the "Pole Spectrometrie Ocean" (PSO) at the University of Brest, France. The parameters used for the Sr isotopic measurement are described in Table 1. The Faraday cups were positioned to collect masses: 82, 83, 84, 85, 86, 87, 88, 90, and 91. Kr interferences on masses 84 and 86 were monitored and corrected by measuring the intensity of ^{82}Kr and ^{83}Kr. The measured $^{82}Kr/^{83}Kr$ ratio was used to correct the natural $^{84}Kr/^{83}Kr$ ratio (4.960835509) and $^{86}Kr/^{83}Kr$ ratio (1.505657093) from the instrumental bias using an exponential law. Once corrected from the

Table 1
MC-ICP-MS Neptune settings for the Sr isotope measurements.

MC-ICP-MS settings	Neptune
RF power (W)	1200
Acceleration potential (V)	10,000
Gas flow rates	
Ar coolant (l/min)	15
Ar auxiliary (l/min)	0.7
Desolvator Apex Q	
Nebulizer pressure (bar)	1.02
Ar sweep gas setting	Not used
N_2 setting	Not used
Solution uptake rate (µl/min)	120
Spray chamber temperature (°C)	120
Desolvator temperature (°C)	2
Analysis parameters	
Number of blocks	1
Number of measurements per block	30
Magnet delay time (s)	0 (static)
Integration time (s)	60 × 4
Typical Sr concentration of samples and standard (ppb)	30
Typical transmission efficiency V/ppm	285