Available online at www.sciencedirect.com

ScienceDirect

Geochimica et Cosmochimica Acta 175 (2016) 282–298

Geochimica et Cosmochimica Acta

www.elsevier.com/locate/gca

Barium isotope abundances in meteorites and their implications for early Solar System evolution

K.R. Bermingham [a,b,*], K. Mezger [a,c], E.E. Scherer [a], M.F. Horan [d], R.W. Carlson [d], D. Upadhyay [a,e], T. Magna [a,f], A. Pack [g]

[a] Institut für Mineralogie, Westfälische Wilhelms-Universität, Corrensstraße 24, 48149 Münster, Germany
[b] Isotope Geochemistry Laboratory, Department of Geology, University of Maryland, College Park, MD 20740, USA
[c] Institut für Geologie, Universität Bern, Baltzerstrasse 1 + 3, 3012 Bern, Switzerland
[d] Department of Terrestrial Magnetism, Carnegie Institution for Science, 5241 Broad Branch Road NW, Washington DC 20015, USA
[e] Department of Geology and Geophysics, Indian Institute of Technology Kharagpur, 721302 Kharagpur, India
[f] Czech Geological Survey, Klárov 3, 11821 Prague 1, Czech Republic
[g] Geowissenschaftliches Zentrum, Georg-August-Universität, Goldschmidtstraße 1, 37077 Göttingen, Germany

Received 17 April 2015; accepted in revised form 4 November 2015; available online 12 November 2015

Abstract

Several nucleosynthetic processes contributed material to the Solar System, but the relative contributions of each process, the timing of their input into the solar nebula, and how well these components were homogenized in the solar nebula remain only partially constrained. The Ba isotope system is particularly useful in addressing these issues because Ba isotopes are synthesized via three nucleosynthetic processes (*s*-, *r*-, *p*-process). In this study, high precision Ba isotope analyses of 22 different whole rock chondrites and achondrites (carbonaceous chondrites, ordinary chondrites, enstatite chondrites, Martian meteorites, and eucrites) were performed to constrain the distribution of Ba isotopes on the regional scale in the Solar System. A melting method using aerodynamic levitation and CO_2-laser heating was used to oxidize SiC, a primary carrier of Ba among presolar grains in carbonaceous chondrites. Destruction of these grains during the fusion process enabled the complete digestion of these samples. The Ba isotope data presented here are thus the first for which complete dissolution of the bulk meteorite samples was certain. Enstatite chondrites, ordinary chondrites, and all achondrites measured here possess Ba isotope compositions that are not resolved from the terrestrial composition. Barium isotope anomalies are evident in most of the carbonaceous chondrites analyzed, but the ^{135}Ba anomalies are generally smaller than previously reported for similarly sized splits of CM2 meteorites. Variation in the size of the ^{135}Ba anomaly is also apparent in fused samples from the same parent body (e.g., CM2 meteorites) and in different pieces from the same meteorite (e.g., Orgueil, CI). Here, we investigate the potential causes of variability in ^{135}Ba, including the contribution of radiogenic ^{135}Ba from the decay of ^{135}Cs and incomplete homogenization of the presolar components on the <0.8 g sample scale.

1. INTRODUCTION

A number of processes can cause isotope ratios in Solar System materials to differ from bulk terrestrial isotope ratios. These processes include (i) radioactive decay (e.g., Reynolds, 1960, 1967; Rowe and Kuroda, 1965), (ii)

* Corresponding author at: Isotope Geochemistry Laboratory, Department of Geology, University of Maryland, College Park, MD 20740, USA. Tel.: +1 (301) 405 2707.
E-mail address: kberming@umd.edu (K.R. Bermingham).

http://dx.doi.org/10.1016/j.gca.2015.11.006
0016-7037/

nuclear reactions following interaction with cosmic rays (e.g., Arnold, 1961), (iii) mass-dependent and -independent isotope fractionation during chemical reactions (e.g., Keesom and van Dijk, 1931; Urey et al., 1932a,b; Thiemens and Heidenreich, 1983; Fujii et al., 2009), and (iv) incomplete mixing in the solar nebula of nucleosynthetic material having exotic isotope abundances inherited from different stellar sources (e.g., presolar grains; Zinner, 1998). The non-solar isotope compositions of individual presolar grains result from nucleosynthetic reactions that were active in their stellar source regions (Burbidge et al., 1957). Ion microprobe (SHRIMP-RG) analyses of the Ba isotopic composition of a SiC-enriched bulk sample (KJB) from Murchison (Ávila et al., 2013) show large anomalies in $\delta^{134}Ba/^{136}Ba$ (140 ± 13), $\delta^{135}Ba/^{136}Ba$ (−423 ± 2), $\delta^{137}Ba/^{136}Ba$ (−284 ± 5), and $\delta^{138}Ba/^{136}Ba$ (−281 ± 23), where $\delta^{i}Ba/^{136}Ba = [(^{i}Ba/^{136}Ba)_{measured}/(^{i}Ba/^{136}Ba)_{solar} - 1] \times 10^3$ (Fig. 1). This isotope signature is generally considered to reflect the *s*-process in Asymptotic Giant Branch (AGB) stars (Savina et al., 2003; Marhas et al., 2007; Ávila et al., 2013). Complementary coupled excesses in ^{135}Ba and ^{137}Ba and have been identified in carbonaceous chondrites and acid leachates (Hidaka et al., 2001, 2003; Andreasen and Sharma, 2007; Carlson et al., 2007; Hidaka and Yoneda, 2011; Qin et al., 2011). The cause of the variations in ^{135}Ba and ^{137}Ba isotope abundances in whole rock meteorites is most frequently attributed to the heterogeneous distribution of presolar grains. This type of isotope anomaly is here termed "nucleosynthetic isotope anomaly" and is the focus of this contribution, which investigates the degree and cause of Ba isotopic heterogeneity in the solar nebula.

Determining the scale of isotope heterogeneity in the solar nebula is an important aspect of cosmochemistry. These data can be used to determine the number and type of supernova contributors to the Solar System, to test the assumption of isotopic homogeneity of the parent isotope that underpins the use of chronometers based on short- and long-lived radioisotopes, and potentially to trace chemical variability in the nebula that might have existed at the time of planetesimal and planet formation. Published ages may be inaccurate if the initial parent/reference isotope ratio (e.g., $^{135}Cs/^{133}Cs$, $^{26}Al/^{27}Al$) had varied throughout the Solar System (e.g., Makide et al., 2011). Such variations in initial abundances could manifest themselves as nucleosynthetic isotope anomalies in bulk meteorite samples. If significant bulk sample nucleosynthetic anomalies were indisputably identified, then the current Solar System chronometers that utilize these isotopes would have to be replaced by regional chronometers that are based on the local abundances of the parent and daughter nuclides.

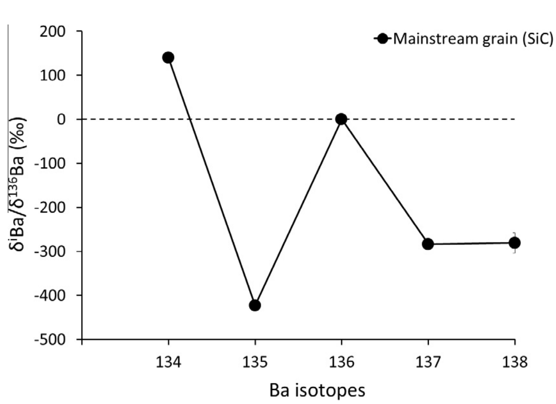

Fig. 1. The Ba isotopic composition of SiC grains from the KJB fraction (Murchison SiC-enriched sample), where $\delta^{i}Ba/^{136}Ba = [(^{i}Ba/^{136}Ba)_{measured}/(^{i}Ba/^{136}Ba)_{solar} - 1] \times 10^3$. Errors are 1$\sigma$ and are smaller than the symbols unless otherwise shown. Data is from Ávila et al. (2013).

How well presolar grains were homogenized throughout the Solar System on the asteroidal- and planetary scales remains unclear. The primary reason for the uncertainty on this point is the difficulty in obtaining *whole rock* chemical analyses of meteorite samples (e.g., carbonaceous chondrites) that contain varying proportions of acid-insoluble components. Regarding the Ba isotope system, the concern is that the documented coupled excesses in ^{135}Ba and ^{137}Ba in carbonaceous chondrites are the result of incomplete dissolution of presolar SiC grains in the bulk sample (e.g., Andreasen and Sharma, 2007; Carlson et al., 2007). Presolar SiC grains display large deficits in ^{135}Ba and ^{137}Ba isotopes and failure to digest part of this component can result in a complementary enrichment pattern in ^{135}Ba and ^{137}Ba, similar in magnitude to the anomalies that are observed in whole rock carbonaceous chondrites. The fact that differentiated and high metamorphic grade meteorites, which no longer contain intact presolar grains, do not display Ba isotope anomalies lends support to this concern.

To evaluate the distribution of Ba isotopes in the Solar System, 22 bulk meteorites from different meteorite classes were analyzed in this study while taking special care to ensure complete dissolution of insoluble phases by using a high temperature laser fusion method. Barium is a moderately refractory element (T_c = 1455 K, $BaTiO_3$; Lodders, 2003; T_c = temperature at which 50% of the element has condensed from the solar nebula) and is a trace element in chondrites (CI = 2.34 ppm; Anders and Grevesse, 1989) with carrier phases including melilite, feldspar, and SiC (estimates of the Ba concentration in individual SiC grains range from ~2 to 10 ppm; Ávila et al., 2013). Barium has seven stable isotopes that sample three different nucleosynthetic processes (*s*-, *r*-, and *p*-processes, Table 1). Low thermal neutron capture cross sections of all Ba isotopes require long irradiation times by cosmic rays to create significant changes in Ba isotope abundances (Table 1; Mughabghab, 2003). There is no reported evidence that indicates secondary nuclear processes produced shifts in Ba isotope abundances in cosmochemical samples. Modification of Ba isotopic composition could also arise from the decay of radioactive species. For example, positive anomalies in ^{135}Ba could result from the decay of the short-lived radioisotope ^{135}Cs ($t_{1/2}$ = 2.3 Myr; Hidaka et al., 2001; Hidaka and Yoneda, 2011, 2013; Bermingham et al., 2014). Lanthanum-138 decays to ^{138}Ba with a half-life of ~105 Gyr (Nakai et al., 1986). Because of the very low natural abundance of ^{138}La (<0.09%) and its long half-life, radiogenic ^{138}Ba is only a very minor component