



Testing the chondrule-rich accretion model for planetary embryos using calcium isotopes



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ABSTRACT

Understanding the composition of raw materials that formed the Earth is a crucial step towards understanding the formation of terrestrial planets and their bulk composition. Calcium is the fifth most abundant element in terrestrial planets and, therefore, is a key element with which to trace planetary composition. However, in order to use Ca isotopes as a tracer of Earth's accretion history, it is first necessary to understand the isotopic behavior of Ca during the earliest stages of planetary formation. Chondrites are some of the oldest materials of the Solar System, and the study of their isotopic composition enables understanding of how and in what conditions the Solar System formed. Here we present Ca isotope data for a suite of bulk chondrites as well as Allende (CV) chondrules. We show that most groups of carbonaceous chondrites (CV, CI, CR and CM) are significantly enriched in the lighter Ca isotopes ($\delta^{44/40}\text{Ca} = +0.1$ to $+0.93\%$) compared with bulk silicate Earth ($\delta^{44/40}\text{Ca} = +1.05 \pm 0.04\%$, Huang et al., 2010) or Mars, while enstatite chondrites are indistinguishable from Earth in Ca isotope composition ($\delta^{44/40}\text{Ca} = +0.91$ to $+1.06\%$). Chondrules from Allende are enriched in the heavier isotopes of Ca compared to the bulk and the matrix of the meteorite ($\delta^{44/40}\text{Ca} = +1.00$ to $+1.21\%$). This implies that Earth and Mars have Ca isotope compositions that are distinct from most carbonaceous chondrites but that may be like chondrules. This Ca isotopic similarity between Earth, Mars, and chondrules is permissive of recent dynamical models of planetary formation that propose a chondrule-rich accretion model for planetary embryos.

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1. Introduction

Chemical and isotopic studies of meteorites are a primary source of information for understanding Solar System formation and evolution, from the nebula phase to the formation of planets. Many meteorites represent materials from the earliest stages of planetary formation and thus their study informs on the origin of terrestrial planets (e.g. Moynier and Fegley, 2015), even if the present-day inventory of meteorites may not necessarily be representative of the accreting materials swept into the planets to form them. The relative chemical abundance of refractory elements in chondrites are close to that of the Sun's photosphere composition and therefore also the solar nebula, making chon-

driles a key source of information about the material that accreted to form the planets (e.g. Palme et al., 2014). Isotopic studies, particularly those using oxygen isotopes, have enabled significant advancement in understanding the relationship between Earth, the Moon and chondrites. Nevertheless, the composition of the bodies that formed the Earth, and the chemical relationship between the Earth and the Moon are still subject to debate (e.g. Dauphas et al., 2014).

Among major elements constituting inner Solar System material, only O and Ca exhibit significant isotopic fractionation among chondrite groups and between chondrites and the bulk silicate earth (BSE) (Clayton and Mayeda, 1999; Simon and DePaolo, 2010; Huang and Jacobsen, 2012; Valdes et al., 2014). Silicon isotopes are mostly fractionated between terrestrial rocks and chondrites (Georg et al., 2007; Savage and Moynier, 2013) and in highly volatile-depleted differentiated meteorites (e.g. Pringle et al., 2014), while other major elements, such as Mg, do not show

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significant fractionation between chondrites and the BSE (e.g. Teng et al., 2010; Bouvier et al., 2013).

Calcium has five stable isotopes, ^{40}Ca (96.94%), ^{42}Ca (0.647%), ^{43}Ca (0.135%), ^{44}Ca (2.086%) and ^{46}Ca (0.004%), and one long-lived radioactive isotope that has such a long half-life that it is essentially stable (^{48}Ca ; 0.187%; half-life = 4.3×10^{19} years). Calcium, despite its relatively high mass, shows large isotopic fractionation up to 1‰ for the $^{44}\text{Ca}/^{40}\text{Ca}$ ratio in the BSE, the Moon, Mars, chondrites and differentiated meteorites (Russel et al., 1978; Niederer and Papanastassiou, 1984; DePaolo, 2004; Simon and DePaolo, 2010; Huang et al., 2012; Valdes et al., 2014; Magna et al., 2015). As Ca is both a lithophile and refractory element (50% condensation $T = 1659$ K, Lodders, 2003), it does not partition into the core, nor it is volatilized during planetary accretion. Therefore its isotopic composition is *a priori* not modified during planetary formation and differentiation of the BSE. These characteristics coupled with the recent development of routine high precision Ca isotopic measurement have led to its use in understanding the composition and origins of the terrestrial planets (Simon and DePaolo, 2010; Huang and Jacobsen, 2012; Valdes et al., 2014). However, high temperature fractionation has been reported for mantle peridotites and co-existing minerals, which may account for some of the variation found in igneous rocks (Huang and Jacobsen, 2012).

Previous Ca isotopic studies showed that different carbonaceous chondrite groups are isotopically distinct and that all groups except the CO chondrites are enriched in the lightest isotopes of Ca, when compared with Earth (Simon and DePaolo, 2010; Huang et al., 2012; Valdes et al., 2014). The origin of the isotopic variations between carbonaceous chondrite groups is puzzling. Although most Ca–Al-rich inclusions (CAIs) are enriched in the lightest isotopes of Ca compared to Earth (Niederer and Papanastassiou, 1984; Huang et al., 2012), the isotopic compositions of bulk carbonaceous chondrites do not correlate with CAI abundances. For example, CI chondrites do not contain CAIs, but are isotopically lighter than CO chondrites, which contain ~10% by volume of CAIs (Zanda et al., 2006). Chondrules are the main carriers of Ca in chondrites (>45% of the total Ca, Rubin, 2011) but, to date, only one high-precision Ca isotope value has been reported for an individual chondrule (from the Allende chondrite) (Simon et al., 2016). This single datum shows that chondrules may be enriched in the heavier isotopes of Ca when compared to the bulk, by about 0.5‰ for the $^{44}\text{Ca}/^{40}\text{Ca}$ ratio.

Recent models have proposed that planetary embryos formed in chondrule-rich regions and that the direct accretion of chondrules would be a viable mechanism to enable rapid growth of planetary embryos up to the size of Mars in the terrestrial planet formation region (Johansen et al., 2015). Since Ca isotopes are not fractionated during planetary accretion, but may be fractionated between matrix, CAIs and chondrules, they can be used to test the model of chondrule accretion. This model predicts that ~90% of the mass of the Earth could be inherited from planetary embryos formed by chondrule accretion (Johansen et al., 2015); therefore, the Ca isotopic composition of Mars and the Earth should be close to the composition of chondrules.

In this study, we present high precision Ca isotopic composition measurements of a variety of terrestrial basalts, bulk chondrites, three individual chondrules and a CAI by multiple-collector inductively-coupled-plasma mass-spectrometry (MC-ICPMS), using the standard bracketing technique, in order to understand the origin of the variations among meteorites and to test the chondrule accretion model for planetary embryos.

2. Materials and methods

2.1. Samples

Samples were chosen to represent a wide range of carbonaceous chondrites: Allende (CV3), Asuka 881595 (CR2), Cold Bokkeveld (CM2), Yamato 980115 (CI1), Orgueil (CI1), Pecora Escarpment (PCA) 02012 (CM2) and PCA 02010 (CM2). Two enstatite chondrites were measured: Indarch (EH4) and Khairpur (EL6). We also measured a mid-ocean ridge basalt (MORB) from the South Mid-Atlantic Ridge (EW9309 10D), two ocean island basalts (OIB) from the Canary Islands (Spain) (Day et al., 2010), one OIB from Hawaii, USA (BHVO-2) and a continental flood basalt from the Colombia River, USA (BCR-2). We also measured a refractory harzburgite xenolith from Lanzarote in the Islands (LZ0604B) with very low loss on ignition (0.04 wt.%) and low Al_2O_3 (0.76 wt.%), and a partially serpentinized harzburgite standard from the USGS (PCC).

In addition to bulk rock samples, one individual CAI from Allende called AB1 was analyzed. It is a fine-grained CAI with a type III REE pattern and has been used for a previous isotopic study of Ba (see data in Moynier et al., 2015). A total of seven chondrules from Allende were analyzed in this study, and these samples were hand-separated using a binocular microscope.

2.2. Sample purification and calcium separation

For the bulk meteorites and chondrules, between 0.8 and 20 mg of sample powder was dissolved in a 3:1 mixture of concentrated hydrofluoric acid and nitric acid (HF/HNO_3) and placed in small Teflon bombs under pressure on a hot plate (120 °C) for two days. The samples were then dried and dissolved again in 6N HCl to destroy fluoride complexes.

The chemical purification follows a modified method described in Valdes et al. (2014). The samples were loaded in 1N HNO_3 on 1.8 mL of pre-cleaned Eichrom DGA resin to separate Ca and Sr from matrix elements. We found that adding a second elution step using 1% H_2O_2 in concentrated HNO_3 generally decreased the width of the Ca elution peak. Hydrogen peroxide (H_2O_2) in solutions was used on the DGA resin by Zhang et al. (2011) to elute titanium (Ti) and separate Ti and Zn from the Zr and Hf fraction. These authors loaded 10 mL of 12N HNO_3 to elute Ca followed by 10 mL of 12N HNO_3 + 1% H_2O_2 to collect Ti. We found that adding a step of 3 mL of 15N HNO_3 + 1% H_2O_2 allowed collection of Ca still attached on the resin. This step includes collection of a minor total fraction of Ti. The inclusion of a minor amount of Ti was deemed acceptable for our measurements, since the interference is on ^{48}Ca , which was not needed for assessing Ca isotopic compositions between planetary materials. This DGA column step was repeated up to three times to ensure clean separation of Ca from matrix elements. We found that the DGA resin can lose capacity after ~10 uses, so we therefore changed the resin after five passes. After the final DGA column pass, the sample residue was dissolved in 1 mL of concentrated HNO_3 to dissolve possible traces of organic matter. Finally, Ca was separated from Sr using 300 μL of Sr-specific resin (Eichrom; 20–50 μm) in Teflon micro-columns following the procedure described in Valdes et al. (2014). One of the difficulties in measuring Ca isotopic compositions is the interference with strontium (Sr). ^{86}Sr and ^{88}Sr are the most abundant Sr isotopes and their doubly charged ions interfere with ^{43}Ca and ^{44}Ca , respectively. Therefore, the Sr had to be removed totally from the Ca cut. The absence of Sr was checked prior to analyzing the samples and is evident from the mass-dependency between the $^{42}\text{Ca}/^{44}\text{Ca}$ and $^{43}\text{Ca}/^{44}\text{Ca}$ (see below). Collections of Ca were then dried down and dissolved in 0.1N HNO_3 for mass spectrometric analysis.

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