



The origin of volatile element depletion in early solar system material: Clues from Zn isotopes in chondrules



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ABSTRACT

Volatile lithophile elements are depleted in the different planetary materials to various degrees, but the origin of these depletions is still debated. Stable isotopes of moderately volatile elements such as Zn can be used to understand the origin of volatile element depletions. Samples with significant volatile element depletions, including the Moon and terrestrial tektites, display heavy Zn isotope compositions (i.e. enrichment of ^{66}Zn vs. ^{64}Zn), consistent with kinetic Zn isotope fractionation during evaporation. However, Luck et al. (2005) found a negative correlation between $\delta^{66}\text{Zn}$ and $1/[\text{Zn}]$ between CI, CM, CO, and CV chondrites, opposite to what would be expected if evaporation caused the Zn abundance variations among chondrite groups.

We have analyzed the Zn isotope composition of multiple samples of the major carbonaceous chondrite classes: CI (1), CM (4), CV (2), CO (4), CB (2), CH (2), CK (4), and CK/CR (1). The bulk chondrites define a negative correlation in a plot of $\delta^{66}\text{Zn}$ vs $1/[\text{Zn}]$, confirming earlier results that Zn abundance variations among carbonaceous chondrites cannot be explained by evaporation. Exceptions are CB and CH chondrites, which display Zn systematics consistent with a collisional formation mechanism that created enrichment in heavy Zn isotopes relative to the trend defined by CI–CK.

We further report Zn isotope analyses of chondrite components, including chondrules from Allende (CV3) and Mokoia (CV3), as well as an aliquot of Allende matrix. All chondrules are enriched in light Zn isotopes (~ 500 ppm on $^{66}\text{Zn}/^{64}\text{Zn}$) relative to the bulk, contrary to what would be expected if Zn were depleted during evaporation, on the other hand the matrix has a complementary heavy isotope composition. We report sequential leaching experiments in un-equilibrated ordinary chondrites, which show sulfides are isotopically heavy compared to silicates and the bulk meteorite by ca. +0.65 per mil on $^{66}\text{Zn}/^{64}\text{Zn}$. We suggest isotopically heavy sulfides were removed from either chondrules or their precursors, thereby producing the light Zn isotope enrichments in chondrules.

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1. Introduction

Carbonaceous chondrites (CC) are chemically primitive early Solar System materials that provide important clues toward understanding the origin and evolution of the terrestrial planets. Carbonaceous chondrites consist of assemblages of four components in different proportion: high-temperature condensates in

the form of Calcium Aluminum-rich Inclusions (CAIs), roughly spherical small igneous objects known as chondrules, metallic FeNi and sulfides, and a fine-grained matrix (Krot et al., 2009; Scott and Krot, 2014). Carbonaceous chondrites are undifferentiated meteorites that are relatively volatile-rich and did not experience high-temperature processing sufficient to result in melting, but they do show evidence of variable degrees of aqueous alteration and thermal metamorphism (Scott and Krot, 2014). Although CC are in general considered as chemically primitive, individual carbonaceous chondrite classes exhibit distinct patterns in volatile element (here “volatile” refers to those elements with a

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50% condensation temperature, T_c , between 250 K and 1250 K, Lodders, 2003) abundances relative to solar composition (Palme et al., 2014a). The CI-type CC are believed to be the closest in composition to the solar nebula due to elemental abundances that closely match the composition of the solar photosphere, with the exception of lithium, the highly volatile elements ($T_c < 250$ K: H, C, N, O), and the noble gases (Palme et al., 2014a). The other CC classes show volatile depletion increasing in the order CI–CM–(CO, CV)–CK (Palme et al., 2014a). The origin of this chemical trend is as yet unresolved. Hypotheses for the volatile element depletion in bulk chondrites include incomplete condensation from the solar nebula (Wasson and Chou, 1974), volatile loss by evaporation during accretion (Ringwood, 1966), or mixing of distinct primordial reservoirs consisting of a volatile-rich CI-like component and a volatile-poor refractory component (Larimer and Anders, 1967; Clayton and Mayeda, 1999; Luck et al., 2003). Chondrules are one proposed carrier of the volatile depletion for the latter case, such that volatile element depletions in bulk meteorites are a signature of distinct chondrule compositions and the different CC classes represent varying mixtures of a compositionally uniform CI-like matrix and compositionally variable chondrules (e.g. Alexander, 2005).

The mechanism of chondrule formation is still debated and it has numerous implications for the origin of volatile elements in the terrestrial planets and for dynamical processes in the early Solar System. Central questions concern the relationship between chondrules and matrix, namely, whether these two chondritic components formed together in the same region of the solar nebula or formed in separate reservoirs and were later combined (Zanda et al., 2006; Hezel and Palme, 2008, 2010; Palme et al., 2015 and references therein). The formation of the first solid materials is therefore directly linked to conditions in the solar environment during early planetary formation and can support or contradict models of Solar System formation. The difficulty lies in reconciling the contrasting formation conditions of CAIs (high temperatures, variable isotopic reservoirs, short formation timescale several Myr before incorporation into chondrites), chondrules (moderately high temperatures, episodic formation likely at higher total pressures), and matrix (lesser thermal processing, volatile-rich) with the incorporation of all three components into a coherent aggregate at a particular time and place in the protoplanetary disk (Krot et al., 2009).

Proposed chondrule formation models that attempt to account for these constraints include the flash heating of chondrule precursors by a shockwave (Connolly and Love, 1998; Ciesla and Hood, 2002; Desch and Connolly, 2002; Connolly and Desch, 2004) or formation in plumes generated by impacts (Asphaug et al., 2011). The flash heating model reproduces the chemical relationships between chondritic components, including the thermal history of chondrules, chondrule–matrix complementarity, and episodic chondrule formation (Asphaug et al., 2011). On the other hand, the impact origin would predict a high gas pressure and dust density that would explain the retention of volatile elements in chondrules. Alternatively, the x-wind model (Shu et al., 1996) is based on astronomical observations of extra-solar systems and theoretical modeling of stellar magnetic fields. It suggests that CAIs and chondrules formed close to the young sun and were later transported radially outward in the disk where they mixed with thermally unprocessed matrix material. This model accounts for the contrasting thermal history between chondrules/CAIs and matrix but fails to explain genetic relationships between chondrules and matrix.

The study of zinc (Zn) isotopes in chondrites and their components has the potential to investigate the numerous questions outlined above. Zinc is a moderately volatile element ($T_c = 726$ K) with a condensation temperature significantly below that of many other moderately volatile elements, including K ($T_c = 1006$ K), Na

($T_c = 958$ K), and Rb ($T_c = 800$ K) (Lodders, 2003). Further, Zn exhibits both chalcophile and lithophile behavior and, hence, provides information about the history of silicate as well as sulphide phases. Zinc displays significant heavy isotope enrichments coupled with volatile depletion, as observed in lunar basalts and unbrecciated eucrites (Paniello et al., 2012a, 2012b; Kato et al., 2015; Moynier et al., 2017). Impact events have been shown to volatilize Zn and drive the composition of the residue toward isotopically heavier compositions (Moynier et al., 2009a), making Zn a powerful probe of volatility-dependent processes during chondrite and chondrule formation. The pioneer study of Luck et al. (2005) reported a negative correlation between $\delta^{66}\text{Zn}$ and $1/[Zn]$ in bulk CC, such that they are progressively enriched in the light isotopes of Zn in the order CI–CM–(CO, CV). This observation is opposite to the effect expected during Zn isotope fractionation due to evaporation and suggests that the abundance of volatile elements in CC is inherited from pre-accretionary processes, reflecting mixing between at least two isotopically distinct reservoirs. The Zn isotope composition of several CC groups (CK, CB, CH), as well as individual components such as chondrules and matrix, which are the major reservoirs of Zn, have not yet been analyzed. However, these samples may provide critical clues toward further understanding the origin of the volatile element variations among CC.

Here we present Zn isotope data for a comprehensive set of CC, including samples from the CI, CM, CO, CV, CK, CB, and CH classes, to further assess the origin and evolution of volatile elements in the inner Solar System. We also present Zn isotope measurements of individual chondrules from Allende (CV3) and Mokoia (CV3) as well as an aliquot of Allende matrix to investigate the formation of solid materials in the early Solar System and the compositions of the primitive materials that represent the building blocks of the terrestrial planets.

2. Samples and methods

2.1. Samples

The 20 bulk chondrites studied include one CI (Y-980115), four CM (Murchison, Murray, Cold Bokkeveld, LON 94101), two CV (Allende, GRA 06101), four CO (Felix, Ornans, Lancé, Isna), two CB (Gujba, MIL 05082), two CH (PCA 91467, A-881020), four CK (ALH 85002, Karoonda, EET 92002, Maralinga), and one CK/CR (A-881595). Y-980115 is CI-like but has mineralogical characteristics that suggest it experienced thermal metamorphism (King et al., 2015). In addition its high Zn content (337 ppm, see below) is similar to other CI chondrites and support its classification as a CI chondrite. A-881595 is classified as a CR2, but more likely is a CK based on O isotopes (Schrader et al., 2011). The bulk compositions of the selected meteorites represent a range in Zn concentration of more than two orders of magnitude, from ~ 3 ppm to ~ 300 ppm. Since Zn is highly mobile in aqueous fluids, meteorite falls were selected over finds when possible. In addition, three unequilibrated ordinary chondrites (UOC), including Clovis (H3.6), GRA 95208 (H3.7), and ALH 90411 (L3.7), were subjected to a sequential dissolution procedure to isolate and analyze the Zn isotope composition of various Zn carrier phases. Finally, nine individual chondrules (including eight from Allende and one from Mokoia) as well as one matrix-rich aliquot from Allende were extracted and their Zn isotope compositions were analyzed.

2.2. Methods

For bulk chondrites, >500 mg of sample was crushed into a homogeneous powder. Approximately 20–50 mg of each sample powder was dissolved using a mixture of concentrated HF/HNO₃ and heated at 130 °C in PTFE beakers. After evaporation of the

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