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Ruthenium isotope fractionation in protoplanetary cores

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Abstract

Mass-dependent Ru isotope variations ($\delta^{102/99}$ Ru) and Ru concentrations were determined for 35 magmatic iron meteorites from the five major chemical groups (IIAB, IID, IIIAB, IVA, IVB). In addition, four equilibrated ordinary chondrites were analyzed. The IIAB, IIIAB and IVB iron meteorites display increasingly heavier Ru isotopic compositions with decreasing Ru content. Modeling demonstrates that the trends for these three iron groups can be reproduced by the incremental extraction of isotopically lighter Ru into solids, which leads to progressively heavier $\delta^{102/99}$ Ru in the remaining melt. The modeling further shows that the Ru isotopic variations of the IIAB and IIIAB irons are consistent with derivation from parental melts with an ordinary chondrite-like $\delta^{102/99}$ Ru, whereas the IVB irons more likely derive from a melt with heavier $\delta^{102/99}$ Ru. This heavy Ru isotopic composition of the IVB parental melt probably results from high-temperature processing of the IVB precursor material. The Ru isotope systematics of the IID and IVA irons are more complex and show no correlation between $\delta^{102/99}$ Ru and Ru content. Although most samples exhibit heavy Ru isotopic compositions, especially the latecrystallized irons of these groups deviate from the expected fractional crystallization trends. This deviation most likely results from mixing and re-equilibration of early-crystallized solids and late-stage liquids, followed by further fractional crystallization. The mixing might be related to the migration of liquids through a complex network of dendrites or to the overturn of a cumulate inner core, and bears testimony to the complex solidification history of at least some protoplanetary cores. © 2017 The Author(s). Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND license (http:// creativecommons.org/licenses/by-nc-nd/4.0/).

Keywords: Ru isotopes; Magmatic iron meteorites; Mass-dependent isotope fractionation; Fractional crystallization; Core solidification; Highly siderophile elements

1. INTRODUCTION

The formation and subsequent solidification of metallic cores during planetary differentiation is one of the most fundamental processes in the early solar system. For most bodies, this process cannot be studied directly, because the cores are inaccessible and hidden in the deep interior of the planets. However, most iron meteorites are generally thought to represent fragments of the disrupted cores of protoplanetary bodies (e.g. Scott and Wasson, 1975), and recent observations of Fe,Ni-rich bodies in the asteroid belt suggest that M-type asteroids (e.g., 16 Psyche) may repre-

* Corresponding author. *E-mail address:* timo.hopp@wwu.de (T. Hopp). sent such bodies (Matter et al., 2013; Shepard et al., 2017). As such, iron meteorites provide the most direct constraints on the processes of core formation and crystallization in small planetary objects.

Magmatic iron meteorites are distinguished from other iron meteorites in that their chemical compositions are broadly consistent with the effects expected from crystalliquid partitioning during progressive crystallization of metallic magma (e.g. Scott, 1972; Scott and Wasson, 1975). Based on their contents of moderately volatile elements (e.g., Ga, Ge), the magmatic iron meteorites are subdivided into eleven groups, each representing metal from a distinct parent body. Of these, the five major groups (IIAB, IID, IIIAB, IVA, IVB) are the most extensively studied. The chemical trends observed among members of each of

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these groups can generally be accounted for by fractional crystallization of metallic melt combined with equilibrium mixing between solids and melts (e.g. Wasson, 1999; Campbell and Humayun, 2005; Wasson et al., 2007; Walker et al., 2008). Whereas this model works reasonably well for the IIAB, IIIAB and IVB groups, more complex processes seem to be necessary to explain the chemical trends observed for the IID and IVA irons (Wasson and Huber, 2006; Goldstein et al., 2009; McCoy et al., 2011).

Until now, the process of core crystallization has mainly been studied using the chemical composition of iron meteorites combined with modeling fractional crystallization. If the iron meteorite cores solidified in this manner, then there should be systematic mass-dependent isotope variations among the irons from a given group. However, evidence for such systematic isotope variations that correlate with the degree of crystallization is sparse. For instance, Chernonozhkin et al. (2016) observed a correlation of Fe stable isotope signatures with decreasing Ir content for IIIAB irons, suggestive of Fe isotope fractionation between liquid and solid metal during fractional crystallization. In addition, Fe isotope fractionation has been observed between metal and troilite in IIAB and IIIAB iron meteorites (Williams et al., 2006). Finally, variations in the Zn isotopic composition of IIAB and IIIAB irons have been attributed to the segregation of chromites rather than progressive crystallization of metal (Bridgestock et al., 2014). Thus, there is currently little evidence for systematic isotope variations resulting from fractional crystallization of iron meteorite cores, and there is no study that investigated such isotope variations for all the major groups of magmatic irons, including the more complex groups IVA and IID.

We present a systematic study of mass-dependent Ru isotopic variations in a comprehensive set of magmatic iron meteorites, including several samples from each of the five major chemical groups (IIAB, IID, IIIAB, IVA, IVB). Ruthenium is ideally suited to examine isotope fractionation during fractional crystallization of protoplanetary cores, because Ru is compatible in solid metal and is therefore continuously removed from the melt during crystallization. Moreover, Ru does not significantly partition into troilites, and so the interpretation of the Ru isotopic data is not complicated by troilite formation. Finally, Ru is sufficiently abundant in almost all iron meteorites, and its isotopic composition can be analyzed with sufficient precision even for very late crystallized samples. The major objectives of the present study are to assess whether or not there is Ru isotopic fractionation during core crystallization, to evaluate if different iron groups with presumably distinct histories display different Ru isotope systematics, and finally to use the Ru isotopic data to gain new insights into the processes operating during the solidification of protoplanetary cores.

2. SAMPLES AND ANALYTICAL METHODS

2.1. Samples and sample preparation

The samples selected for this study include 35 magmatic iron meteorites from the five major chemical groups (IIAB, IID, IIIAB, IVA, IVB) and four ordinary chondrites (H5, L6). The ordinary chondrite samples were cut from larger slices, and cleaned with SiC abrasives and de-ionized water in an ultrasonic bath. Then the samples were crushed to fine powders in an agate mortar. About 0.5 g of each sample powder was weighed into Carius tubes and spiked with an appropriate amount of ⁹⁸Ru-¹⁰¹Ru double spike. The sample-spike mixtures were digested using 5 ml concentrated HNO₃ and 2.5 ml concentrated HCl inside sealed Carius tubes at 220 °C for 48 h (Shirey and Walker, 1995; Hopp et al., 2016). In ordinary chondrites of petrologic types 5 and 6, Ru is hosted in metals, sulfides and to a minor degree oxides. During Carius tube digestion metals and sulfides are completely dissolved and Ru from oxides (e.g., spinel) is leached. As such, the Carius tube digestion quantitatively assesses the Ru present in the sample. After digestion, the sample solutions were transferred into 50 ml centrifuge tubes and centrifuged for 20 min to separate and remove the un-dissolved silicates. After centrifuging, the solution was transferred into 60 ml Savillex PFA beakers.

The iron meteorite samples were either obtained as small chips or sawn from larger slabs. Weathered material and fusion crust, if present, were removed. All samples were polished and cleaned using SiC abrasives and de-ionized water in an ultrasonic bath. After cleaning, between ~ 50 and \sim 360 mg of each sample was weighed into 60 ml Savillex PFA beakers, spiked with a ⁹⁸Ru-¹⁰¹Ru double spike and digested in 10 ml 6 M HCl at 120 °C on a hot plate. The metals were fully dissolved after $\sim 1-2$ h, and the sample solutions were then cooled down and ~ 10 ml of concentrated HNO₃ was added to produce reverse agua regia. This solution was placed on a hot plate at 120 °C overnight to ensure spike-sample equilibration. To test as to whether full spike-sample equilibration is obtained in this manner, four of the iron meteorites (Henbury, Sikhote Alin, Grant and Ainsworth) were also processed using the Carius tube digestion method described above, where sample and spike were equilibrated in reverse aquia regia at 220 °C for 48 h. The results from the table top and Carius tube digestions are in very good agreement, indicating that full spike-sample equilibration is achieved with the table top digestion (Fig. 1).

After digestion, all sample solutions were dried down at 100 °C on a hot plate and re-dissolved twice with 10 ml of 6 M HCl. This procedure ensures complete removal of HNO_3 from the sample solutions. After conversion into chloride form, the samples were re-dissolved in 10 ml of 0.2 M HCl for further processing on cation exchange columns.

2.2. Chemical separation of Ru

The purification of Ru from the samples followed our established procedures described in Fischer-Gödde et al. (2015) and Hopp et al. (2016). In brief, Ru together with other highly siderophile elements (HSE) and some other trace elements (e.g., Mo) was separated from the major sample matrix using cation exchange columns filled with 10 ml of pre-cleaned BioRad AG50 W-X8 (100–200 mesh) resin. Samples were loaded onto the column in 10 ml 0.2

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