Earth and Planetary Science Letters 490 (2018) 31–39

Contents lists available at ScienceDirect

Earth and Planetary Science Letters

www.elsevier.com/locate/epsl

Fe isotope composition of bulk chondrules from Murchison (CM2): Constraints for parent body alteration, nebula processes and chondrule-matrix complementarity

Dominik C. Hezel [a,b,c,*], Johanna S. Wilden [a], Daniel Becker [a], Sonja Steinbach [d], Frank Wombacher [a,c], Markus Harak [a]

[a] University of Cologne, Department of Geology and Mineralogy, Zülpicher Str. 49b, 50674 Köln, Germany
[b] Natural History Museum, Department of Mineralogy, Cromwell Road, SW7 5BD, London, UK
[c] Steinmann-Institut, Poppelsdorfer Schloss, Meckenheimer Allee 169, 53115 Bonn, Germany
[d] Deutsches Zentrum für Luft- und Raumfahrt (DLR), Institut für Materialphysik im Weltraum, Linder Höhe, 51147 Köln, Germany

ARTICLE INFO

Article history:
Received 25 November 2017
Received in revised form 29 January 2018
Accepted 8 March 2018
Available online xxxx
Editor: F. Moynier

Keywords:
chondrules
matrix
Fe isotopes
tomography
chondrule formation
complementarity

ABSTRACT

Chondrules are a major constituent of primitive meteorites. The formation of chondrules is one of the most elusive problems in cosmochemistry. We use Fe isotope compositions of chondrules and bulk chondrites to constrain the conditions of chondrule formation. Iron isotope compositions of bulk chondrules are so far only known from few studies on CV and some ordinary chondrites. We studied 37 chondrules from the CM chondrite Murchison. This is particularly challenging, as CM chondrites contain the smallest chondrules of all chondrite groups, except for CH chondrites. Bulk chondrules have δ^{56}Fe between −0.62 and +0.24‰ relative to the IRMM-014 standard. Bulk Murchison has as all chondrites a δ^{56}Fe of 0.00‰ within error. The δ^{56}Fe distribution of the Murchison chondrule population is continuous and close to normal. The width of the δ^{56}Fe distribution is narrower than that of the Allende chondrule population. Opaque modal abundances in Murchison chondrules is in about 67% of the chondrules close to 0 vol.%, and in 33% typically up to 6.5 vol.%. Chondrule Al/Mg and Fe/Mg ratios are sub-chondritic, while bulk Murchison has chondritic ratios. We suggest that the variable bulk chondrule Fe isotope compositions were established during evaporation and recondensation prior to accretion in the Murchison parent body. This range in isotope composition was likely reduced during aqueous alteration on the parent body. Murchison has a chondritic Fe isotope composition and a number of chondritic element ratios. Chondrules, however, have variable Fe isotope compositions and chondrules and matrix have complementary Al/Mg and Fe/Mg ratios. In combination, this supports the idea that chondrules and matrix formed from a single reservoir and were then accreted in the parent body. The formation in a single region also explains the compositional distribution of the chondrule population in Murchison.

1. Introduction

Chondritic meteorites (chondrites) are “cosmic conglomerates” (cf. Brearley and Krot, 2012) and mainly consist of chondrules plus matrix (up to >90 vol.%), and minor amounts of Ca, Al-rich inclusions and opaque phases (mainly metal, sulfide and magnetite). Chondrules are roundish, typically tens of µm to mm in diameter sized, igneous objects that formed during brief high-temperature events in the early solar system. Many models of chondrule formation have been proposed over the past decades. The difficulty to identify the chondrule forming process results primarily from insufficient constraints on chondrule formation and contradictions between these constraints. Two of the most critical constraints regarding chondrule formation currently discussed are: (i) how to explain the compositional distributions of chondrule populations in individual meteorites, and (ii) did chondrules and matrix form in the same or in different regions of the protoplanetary disk and were subsequently transported and aggregated into a single parent body. Further, it is important to know whether chondrule formation conditions are similar or different between groups or even individual meteorites of the same group (e.g., Ciesla, 2005; Zanda et al., 2006; Hezel et al., 2006; Hezel and Palme, 2008, 2010; Alexander and Ebel, 2012; Jones, 2012; Palme et al., 2014b, 2015; Becker et al., 2015; Goldberg et al., 2015; Olsen et al., 2016).

* Corresponding author at: University of Cologne, Department of Geology and Mineralogy, Zülpicher Str. 49b, 50674 Köln, Germany.
E-mail address: dominik.hezel@uni-koeln.de (D.C. Hezel).

https://doi.org/10.1016/j.epsl.2018.03.013
0012-821X/

The distribution of stable Fe isotopes in the chondrule population of individual chondrites record the conditions during chondrule formation, as well as information about the region in the protoplanetary disk where chondrules formed. So far, Fe isotope compositions of chondrules and matrix have only been reported for few meteorites: for the CV chondrites Allende, Mokoia and Grosnaja (Mullane et al., 2005; Hezel et al., 2010) and various ordinary chondrites (Kehm et al., 2003; Mullane et al., 2005; Needham et al., 2009). Chondrules have large stable Fe isotope variations, unlike their host bulk chondrites, which are highly homogeneous and indistinguishable from each other (Zhu et al., 2001; Kehm et al., 2003; Poitrasson et al., 2004; Wang et al., 2013; Mullane et al., 2005; Needham et al., 2009; Hezel et al., 2010). Stable isotope variations are expressed in parts per thousand deviation from a standard, and in chondrules the δ^{56}Fe ranges from -1.33 to $+0.65$ ‰. For comparison, most igneous terrestrial rocks (e.g., peridotites and abyssal basalts) range between -0.43 and $+0.18$ ‰ (e.g., Weyer and Ionov, 2007; Teng et al., 2013).

A number of processes have been suggested to explain the observed δ^{56}Fe variations in chondrules: Needham et al. (2009) found that the δ^{56}Fe distribution of chondrite chondrule populations becomes narrower with increasing petrologic type of their host chondrites. Hence, parent body metamorphism redistributes Fe, and thereby the chondrule δ^{56}Fe compositions decrease towards the host chondrite's δ^{56}Fe compositions. Mullane et al. (2005) suggested that a combination of high-temperature nebula events and subsequent metasomatic processes on the parent body produced the observed δ^{56}Fe distribution in Allende and Chainpur chondrule populations. Bouvier et al. (2013) reached a similar conclusion by using stable Mg isotope compositions of bulk chondrules from the CM chondrites Murchison and Murray. Finally, Hezel et al. (2010) concluded that the dominant process for the δ^{56}Fe distribution in the Allende chondrule population most likely was evaporation and recondensation in the protoplanetary disk. Other possibilities to explain these isotope distributions could be compositionally heterogeneous chondrule precursor grains or formation of chondrules in compositionally different regions in the protoplanetary disk with subsequent transport and mixing into their parent bodies.

Bulk chondrule Fe isotope data from carbonaceous chondrites so far only exist for CV chondrites. Here we study chondrules from the CM chondrite Murchison to (i) expand our knowledge of bulk chondrule Fe isotope distributions to a second carbonaceous chondrite group, (ii) compare our results to the CV chondrite chondrule populations and (iii) to provide constraints for the formation and origin of chondrules.

2. Methods

2.1. Sample selection

A total of ~2 g Murchison was wrapped in weighing paper and gently crushed using a small hammer. Individual chondrules were separated under a binocular with ceramic tweezers to avoid any contamination from metal. In rare cases, matrix adhering to chondrules was removed using a second set of ceramic tweezers. Separated chondrules were stored individually in small, labelled gelatine capsules.

2.2. 3D μ-tomography

The method of 3D X-ray computed tomography allows non-destructive, high spatial and contrast resolution imaging of the volume fraction of different chondrule phases in three dimensions. We used different mounts for tomography measurements. In a first round of measurements, the chondrules remained in their gelatine capsules, which were placed in a test tube. This approach was only partly successful, as some of the chondrules moved during the scans. For the next round, we built a sample holder made of X-ray transparent PVC that can take and measure up to 7 chondrules per scan. The chondrules are fixed in small holes with cotton in the top of the holder. With this setup, none of the chondrules moved during the CT-scans. The holders were mounted on the rotating table of a Nanotom® 160NF (Phoenix | X-ray, Germany) at the German Aerospace Center DLR, Cologne, Germany. The measurements were conducted with a tube voltage set to 100 kV and the current set to 100 µA. A total of 1000 projections were acquired, as the sample was rotated through 360° in incremental steps of 0.36°, with each projection taking 2 s. Dark current noise and spatial heterogeneity of the X-ray beam were corrected by dark current subtraction and division of the direct beam images. The voxel resolution was 7.5 µm. The reconstruction of the 16-bit images was performed with datos | x-reconstruction software (GE Sensing & Inspection GmbH). Quantitative results were extracted from subsets of the tomographic data (image stacks) using ImageJ.

2.3. Sample preparation and dissolution

Individual chondrules were dissolved in closed Savillex teflon beakers for at least 12 h at >120 °C in a 2:1 mixture of concentrated HF and concentrated HNO_3. The solutions were dried down at 90 °C and the residues redissolved in concentrated HCl for at least 12 h at >120 °C. This second step was repeated. No residual solids were found at this stage. A total of 7 g Murchison was powdered in an agate mortar, of which ~50 mg were used for bulk Murchison analyses. Further, two aliquots of a few milligrammes of Jbilet Winselwan (CM) powder (Friend et al., 2018, pers. comm.) as well as two aliquots of a few milligrammes of Smithsonian Allende (CV) reference powder were digested with the same procedure used for the chondrules (cf. Hezel et al., 2015).

2.4. Element analyses

A set of 6 elements (Al, Mg, Fe, Ni, Cr, Ca) was measured by means of Optical Emission Spectrometry (ICP-OES), using a Spectro Arcos OES in the University of Cologne facilities. Detection limits for all elements are in the range of 10 ppb or less, and errors are <5 rel.%. As CM chondrules are the smallest – except for CH – we had to preserve as much Fe as possible for isotope analyses, and diluted the solution by at least 1:100. Therefore Cr and other elements with low concentrations are often below or close to the detection limit. This dilution might also introduce some uncertainty to the element concentrations. Only element ratios are given, as we did not determine precise chondrule weights, which would be required to recalculate chondrule element concentrations from the OES measurements.

2.5. Fe separation and mass spectrometry

Chromatographic separation of Fe was performed following published methods (Dauphas et al., 2004) using AG1-X4 anion exchange resin. ICP-OES analyses confirmed that complete recovery of Fe was achieved. Sample to procedural blank ratios typically were at least 1000:1, and in rare cases at least 100:1, thereby introducing either no or only a negligible error.

The Fe isotope compositions were measured at the joint Cologne–Bonn laboratory, using a Thermo Finnigan Neptune MC-ICP-MS, in conjunction with a Micromist nebuliser and a dual cyclonic-Scott type glass spray chamber for sample introduction. The analyses were carried out in medium resolution mode. This configuration provided sufficient transmission to allow routine analyses of 1 ppm Fe sample solutions. The δ^{56}Fe and δ^{57}Fe notation refers to the permil deviation of the ^{56}Fe/^{54}Fe and ^{57}Fe/^{54}Fe