Earth and Planetary Science Letters 423 (2015) 13–23

Contents lists available at ScienceDirect

Earth and Planetary Science Letters

www.elsevier.com/locate/epsl

Early cosmic ray irradiation of chondrules and prolonged accretion of primitive meteorites

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ARTICLE INFO

Article history:
Received 4 February 2015
Received in revised form 9 April 2015
Accepted 13 April 2015
Available online 29 April 2015
Editor: B. Marty

Keywords:
chondrules
solar nebula
cosmogenic noble gases
pre-irradiation
galactic cosmic rays
solar cosmic rays

ABSTRACT

Chondrules, together with Ca-Al-rich inclusions (CAIs) and matrix, are the major constituents of primitive meteorites. It is clear that chondrules formed as molten objects and the conditions under which this happened seem well constrained. Partially overlapping in age, but mostly ∼2–3 million years younger than the CAIs, they appear to have formed over an extended period of time (e.g., Kita et al., 2013). We have analyzed chondrules in two highly primitive CR3 meteorites, QUE 99177 and MET 00426, and find that they contain highly variable amounts of noble gases produced by irradiation with cosmic rays. The lack of implanted solar wind and the composition of the cosmogenic component in QUE 99177 chondrules argue against irradiation in a parent body regolith, which leaves irradiation in the early solar system as the most likely explanation. The cosmogenic composition also points to irradiation primarily by galactic cosmic rays (GCR), not solar cosmic rays (SCR), i.e. not by an active early sun. To allow effective production of cosmogenic isotopes by GCR, but not SCR, this should have happened rather "late" in a largely, but not completely, dust-free environment. Our results support the suggestion that chondrules formed as free-floating objects in the solar nebula; also consistent with the noble gas data is pre-irradiation in small (∼dm-size) aggregates that broke up before or during accretion to the CR parent body. In both cases, chondrules spent an extended period of time before incorporation into the most primitive meteorite parent bodies, which puts constraints on accretion time scales.

1. Introduction

There has been remarkable progress during recent years in dating the formation of the first solids formed in the Solar System, CAIs and chondrules, both using long-lived chronometers (Pb–Pb) as well as short-lived extinct nuclides, from which a largely consistent picture has emerged (e.g., Connelly et al., 2012; Kita et al., 2013). The parent bodies of iron meteorites must have accreted rapidly as shown by the ^{182}Hf–^{182}W short-lived chronometer, which dates core formation (e.g., Kleine et al., 2009; Wittig et al., 2013; Kruijer et al., 2014), but more open is the question of how long it took for the first solids to accrete to those planetesimals that are the parent bodies for primitive chondritic meteorites. Dating of aqueous alteration products may put an upper limit of a few million years to this time interval (e.g., Hoppe et al., 2008; Trinquier et al., 2008), since it is generally assumed to have taken place on the parent bodies. As early heating, differentiation, and thermal metamorphism was induced by thermal effects mostly from ^{26}Al decay, thermal modeling is another tool that sets a lower limit of chondrite parent body accretion of at least 2 Ma after CAIs (e.g., Henke et al., 2012, 2013; Trieloff et al., 2003).

Unless shielded after formation in a dust-rich region, the early formed solids must have been exposed to cosmic radiation, either to solar cosmic rays (SCR) and/or galactic cosmic rays (GCR), and the presence of cosmic ray produced nuclides should provide another, more direct measure. Evidence of an early irradiation by an active Sun appears to be recorded in CAIs in the form of now extinct ^{10}Be (e.g., McKeegan et al., 2000; Krot et al., 2009). Similarly sensitive to the effects of production by cosmic rays as short-lived radionuclides are the noble gases, due to their low natural abundance in rocky material. Consequently, there have been several recent searches, using noble gases, for "pre-irradiation" of chondrules (Polnau et al., 1999, 2001; Eugster et al., 2007; Das and Murty, 2009; Roth et al., 2011).

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http://dx.doi.org/10.1016/j.epsl.2015.04.024
0012-821X/

Here "pre-irradiation" is identified by means of an excess of cosmogenic isotopes relative to what has been produced during the recent space travel of the meteorites from their parent body to Earth, due to an additional episode of earlier production. There are various ways in which this may have happened. The meteorite (rock) may have been exposed while residing near the surface of its parent body (e.g., Begemann et al., 1985). It may contain clasts that have been pre-irradiated there (e.g., Wieler et al., 1989a) or, if foreign, during travel from or on an earlier parent body (Wieler et al., 1989b). In these cases, if there is a sufficient time span between the early and the "recent" cosmic ray exposure (and the latter was not too long), this may be recognized by differences in the results obtained from cosmogenic radionuclides vs. stable cosmogenic products.

In a different situation even smaller units like individual grains (Hohenberg et al., 1990), although this interpretation has been challenged (Wieler et al., 1989b, 1990), or individual components such as chondrules (of interest here) or Ca-Al-rich inclusions (CAIs) may have been pre-irradiated. Again, one setting where this may have happened is a parent body regolith, and the presence of implanted solar wind is usually taken as a sign of parent body surface residence. In fact, solar wind is abundantly present in CM2 Murchison and CR2 NWA 852, where clear evidence for pre-irradiation of chondrules has been found (Roth et al., 2011; Beyersdorf-Kuis, 2014), which thus can confidently be ascribed to precompaction irradiation on the parent body.

The situation is less clear concerning the few positive observations tentatively ascribed to early solar system pre-irradiation (Polnau et al., 1999, 2001; Eugster et al., 2007; Das and Murty, 2009). There are several reasons and pitfalls involved in these studies and the differences in "nominal" cosmic ray exposure time tentatively ascribed to pre-irradiation by GCR are either small (<1 Ma) or highly uncertain. The report by Das et al. (2012), on the other hand, who analyzed individual grains from within chondrules and claimed to have found evidence for SCR irradiation, is compromised by the use of incorrect target element abundances. When corrected, the claimed effects disappear (Ott et al., 2013). In our approach we avoided the most important problems. In particular, the abundances of target elements were determined by Instrumental Neutron Activation Analysis (INAA) on exactly the same material subsequently used for noble gas analysis, thus avoiding effects of sample inhomogeneity on cosmogenic production rates. Moreover we concentrated on highly primitive meteorites, which are least likely to have lost memory of nebular events.

Results reported here are for chondrules from two CR meteorites, which based on various criteria including the abundance of presolar silicates (Floss and Stadermann, 2009) are among the most primitive ones in our collections, QUE 99177 and MET 00426. Having hardly been affected by thermal and aqueous metamorphism, they have been classified as CR3.0 (Abreu and Brearley, 2010) or CR2.8 (Harju et al., 2014). Data obtained on several others (Beyersdorf-Kuis, 2014) will be reported separately. Among these is the Allende meteorite, for which we find no evidence for pre-irradiation, in agreement with previous studies (Das and Murty, 2009; Roth et al., 2011).

Most intriguing are the results for QUE 99177, which shows no detectable implanted solar wind and where, compared to previous reports of "sporadic" (Das and Murty, 2009) occurrences of pre-compaction irradiation assuming an early solar system setting (Polnau et al., 1999, 2001; Eugster et al., 2007; Das and Murty, 2009), effects are much more clear-cut. We believe that these results present the strongest evidence so far that pre-irradiation in the early solar system did indeed take place, a fact which is bolstered by the composition of the cosmogenically produced noble gas component. In spite of the presence of solar wind gas, the similarity to QUE 99177 in many aspects (Floss and Stadermann, 2009; Abreu and Brearley, 2010), in combination with the similarity of the cosmic ray effects, suggests the same for MET 00426. If, after all, in the unlikely case that the observed unique kind of pre-irradiation happened on the parent body instead, this naturally has interesting consequences for dynamical processes in its regolith.

2. Materials and methods

2.1. Samples; separation of chondrules

QUE 99177 and MET 00426 are highly primitive carbonaceous chondrites classified as CR3.0 (Abreu and Brearley, 2010) or CR2.8 (Harju et al., 2014). They are finds collected in Antarctica and about 300 mg of each were obtained for this study from the Meteorite Working Group.

In previous work on chondrules various types of mechanical separation methods (Polnau et al., 1999, 2001; Eugster et al., 2007) as well as the freeze–thaw technique (Das and Murty, 2009; Roth et al., 2011) have been used, the choice and details dictated by mechanical properties like the friability of the host meteorites. In our case, after initial microscopic examination, we applied the freeze–thaw technique largely following the method of Grossman (2010).

In brief, samples in Teflon containers were immersed into distilled water and were evacuated in order to suck air out of pore spaces and allow water to penetrate. This was followed by dipping the vials into liquid nitrogen, which caused the water to freeze and expand, thus leading to disaggregation of the sample. After thawing the sample by warming in a water bath, the cycle was repeated. Ultrasonic agitation to speed up the process was added after each ten cycles. Supernatant finely dispersed material was decanted and dried and constitutes our "matrix" material, while chondrules were hand-picked under a binocular microscope from the remaining material. Altogether we applied 60×10 cycles.

It is not clear how well the composition of our "matrix" corresponds to that of true matrix. Fine-grained material in QUE 99177 and MET 00426 occurs in interchondrule matrix, fine-grained rims and dark inclusions (Abreu and Brearley, 2010). Interchondrule matrix is the most abundant among these (Abreu and Brearley, 2010), so we assume that it also dominates our "matrix". Material from broken-up chondrules and CAIs can be expected to be present as well.

2.2. Neutron activation analysis

Instrumental neutron activation analyses of the chondrules and the matrix material used for later noble gas analyses were performed at the Australian Nuclear Science and Technology Organisation (ANSTO) in Kirrawee, DC, Australia, using the k_0 method. Details are given in Bennett (2008) and Murrie et al. (2013). To test the reliability and reproducibility we also analyzed various small samples of finely and more coarsely powdered material from the L chondrite ALH 81032. The obtained results closely agree with average concentrations in L chondrites (Wasson and Kallemeyn, 1988). Small observed variations are most likely due to a small compositional heterogeneity showing up if small sample sizes are used and no special effort is made to produce true aliquots. For our specific chondrules irradiation times were adjusted to sample size and ranged from ∼4 min for the rather large chondrules #6 from both meteorites to 15 min for the majority of the chondrules. The flux density was $\sim 2 \times 10^{13}$ neutrons $cm^{-2}\,s^{-1}$, and the matrix samples were irradiated for one minute only each. Table 1 provides the individual neutron doses together with the results of the INAA analyses.