



# Oxygen isotopic diversity of chondrule precursors and the nebular origin of chondrules

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## ABSTRACT

FeO-poor (type I) porphyritic chondrules formed by incomplete melting of solid dust precursors via a yet-elusive mechanism. Two settings are generally considered for their formation: (i) a nebular setting where primordial solids were melted, e.g. by shock waves propagating through the gas and (ii) a collisional planetary setting. Here we report a method combining high-current electron microprobe X-ray mapping and quantitative measurements to determine the chemical characteristics of relict olivine grains inherited from chondrule precursors. We find that these olivine crystals are Ca–Al–Ti-poor relative to host olivine crystals. Their variable  $\Delta^{17}\text{O}$ , even in individual chondrule, is inconsistent with derivation from planetary interiors as previously argued from  $120^\circ$  triple junctions also exhibited by the chondrules studied herein. This indicates that chondrule precursors correspond to solid nebular condensates formed under changing physical conditions.

We propose that porphyritic chondrules formed during gas-assisted melting of nebular condensates comprising relict olivine grains with varying  $\Delta^{17}\text{O}$  values and Ca–Al–Ti-rich minerals such as those observed within amoeboid olivine aggregates. Incomplete melting of chondrule precursors produced Ca–Al–Ti-rich melts (CAT-melts), allowing subsequent crystallization of Ca–Al–Ti-rich host olivine crystals via epitaxial growth on relict olivine grains. Incoming MgO and SiO from the gas phase induced (i) the dilution of CAT-melts, as attested by the positive Al–Ti correlation observed in chondrule olivine crystals, and (ii) buffering of the O-isotope compositions of chondrules, as recorded by the constant  $\Delta^{17}\text{O}$  values of host olivine grains. The O-isotopic compositions of host olivine grains are chondrule-specific, suggesting that chondrules formed in an array of environments of the protoplanetary disk with different  $\Delta^{17}\text{O}$  values, possibly due to variable solid/gas mixing ratios.

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## 1. Introduction

Chondrules (millimeter-sized igneous spheroids containing silicates, metal, sulfides, and glass) are the major high-temperature components of primitive meteorites (chondrites), suggesting that most inner solar system materials were affected by their formation. However, the underlying mechanism(s) of their formation remains a mystery and diverse scenarios are debated in contemporaneous literature. A key clue to their origin would be the identification of the precursor material that was melted to form chondrules. In this effort, cosmochemists may find help in the incomplete melt-

ing of most chondrules, as evidenced by their widespread porphyritic texture (Hewins et al., 2005). Indeed, relict grains inherited from chondrule precursors are identifiable. Forsteritic grains in high-FeO (type II) porphyritic chondrules were presumably inherited from precursors formed in more reducing conditions than their current host (Nagahara, 1981; Scott and Jones, 1990). Conversely, “dusty” grains, i.e., crystals speckled with Fe-rich metal beads, in low-FeO (type I) chondrules are commonly attributed to oxidized precursors that underwent reduction during formation of their host chondrules (Lemelle et al., 2001; Leroux et al., 2003; Nagahara, 1981; Rambaldi, 1981). These examples represent only a fraction of the existing relicts. In type I chondrules—the dominant type in carbonaceous chondrites and hence the main asteroid belt—relict grains from reduced precursors are not so easily recognizable.

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Oxygen isotopic systematics may help to identify such relicts (Kimura et al., 2011; Kunihiro et al., 2004, 2005; Rudraswami et al., 2011; Schrader et al., 2013, 2015; Tenner et al., 2013, 2015; Ushikubo et al., 2012). Indeed significant isotopic diversity is observed among solar system materials, from  $^{16}\text{O}$ -rich refractory inclusions (calcium–aluminum-rich inclusions (CAIs), amoeboid olivine aggregates (AOA)) to  $^{16}\text{O}$ -poorer chondrules (Clayton, 2003). Type I chondrules exhibit significant variability and define a broad line in the oxygen three-isotope diagram that is not specific to chondrules of a given chondrite or to particular chondrite type and known as the primitive chondrule minerals (PCM) line (Ushikubo et al., 2012). The underlying  $^{16}\text{O}$  variability could result from (i) physical mixing of grains of different origins within the solid chondrule precursors (Hezel and Palme, 2007; Tenner et al., 2015) and/or (ii) high-temperature exchanges between the chondrule melt and the surrounding gas (Marrocchi and Chaussidon, 2015). Secondary ion mass spectrometer (SIMS) analyses of olivines within a single chondrule typically reveal homogeneous oxygen isotopic compositions (Chaussidon et al., 2008; Tenner et al., 2015). Yet, in some chondrules, some olivines are  $^{16}\text{O}$ -enriched compared to their neighbors (Kunihiro et al., 2005; Rudraswami et al., 2011): these are generally considered to be relict grains, even if they are not petrographically manifest (Ushikubo et al., 2012).

Despite these additions to the known inventory of relict grains, their origin remains unclear. Relict olivine grains could correspond to (i) early condensates from the gas of the solar protoplanetary disk (Cohen et al., 2004; Jacquet and Marrocchi, 2017; Russell et al., 2005; Yurimoto and Wasson, 2002), (ii) collisional debris from early-generation planetesimals (Libourel and Chaussidon, 2011; Libourel and Krot, 2007) and/or (iii) earlier generations of chondrules (e.g., Ruzicka et al., 2007). Progress on the identification of the origin of relict olivine grains is frustrated by our poor understanding of their abundance, distribution and specific chemical compositions (Pack et al., 2005, 2004; Ruzicka et al., 2007). Among isotopic studies, only a few *in situ* measurements of olivine are generally performed within a single chondrule (usually 2–10; Rudraswami et al., 2011), leading to an underestimate of the abundance of relict olivine grains (and relict-bearing chondrules) and a lack of knowledge of their isotopic characteristics. Furthermore, there are no clear major-element signatures and/or textural features that facilitate the recognition of relict olivine grains (Jones et al., 2004; Ushikubo et al., 2012). Consequently, the message carried by relict olivine grains remains difficult to decipher, however fundamental it would be to understand the origin of chondrules.

Here we report a new method combining high-resolution X-ray maps, electron microprobe analyses, and SIMS oxygen isotope measurements to quantitatively assess for the first time the nature of relict olivine grains in type I chondrules. This method was successfully applied to type I porphyritic chondrules in the CM-related ungrouped chondrite Northwest Africa (NWA) 5958 (Jacquet et al., 2016) and provides new information on the formation conditions of the first solids of the solar system.

## 2. Material and methods

NWA 5958 is a carbonaceous chondrite found in the Moroccan desert in 2009 that shares numerous similarities with the CM chondrite group. This meteorite shows limited terrestrial weathering and a low degree of aqueous alteration (Jacquet et al., 2016). Its bulk oxygen isotopic composition ( $\Delta^{17}\text{O} = -4.3\text{‰}$ ) is more  $^{16}\text{O}$ -rich than all CM chondrites, further supporting a limited alteration episode (Marrocchi et al., 2018; Verdier-Paoletti et al., 2017). As previous O isotope studies have revealed CO chondrules to be richest in relicts (Tenner et al., *in press*), this meteorite, as a mem-

ber of the CM-CO clan, was a sample of choice to increase their chances of discovery.

We surveyed all type I chondrules in thick section NWA 5958-1 (from the Muséum national d'Histoire naturelle collection, Paris, France) of NWA 5958. The X-ray map of NWA5981-1 was reported elsewhere (Fig. 1 of Jacquet et al., 2016). Chondrules were examined microscopically in reflected light. Scanning electron microscope observations were performed at CRPG using a JEOL JSM-6510 with 3 nA primary beam at 15 kV. Among 32 Mg-rich porphyritic Type I chondrules examined, we selected 3 olivine-rich chondrules (Fig. 1, 2, S1–S3). High-resolution X-ray element distribution maps were performed at the Institut des Sciences de la Terre (ISTerre, Grenoble, France), using a JEOL JXA-8230 electron microprobe analyzer (EMPA) equipped with five wavelength-dispersive spectrometers (WDS) and one silicon drift detector energy-dispersive spectrometer (EDS; Batanova et al., 2015). Analysis were performed with an acceleration voltage of 20 kV, beam current of 500 nA, 1.5  $\mu\text{m}$  step size, and dwell time of 500 ms. Al, Ca, Cr, Mn and Ti were measured by WDS while Fe, Si and Mg were measured by EDS. Each map last 24 hours followed by 8 hours of quantitative analyses. Detection limits were below 40 ppm for Al, Ca, Ti, Cr and Mn as estimated from minimum differences between the average concentrations of the visually distinct homogeneous zones and standard deviations of concentration of homogeneous zones. These X-ray maps are semi-quantitative, as the background was not quantified by performing similar off-peak maps. However, quantitative analyses of all olivine grains large enough to be isotopically characterized by SIMS were then performed with the following conditions: accelerating voltage 20 kV, probe current 900 nA, beam diameter 1  $\mu\text{m}$ , and 320 s total peak/background counting time. Such a high current and long counting time allow very low detection limits estimated to be 10 ppm for Al, Ca, Ti, Cr and Mn. The ZAF software was used for matrix corrections (see Batanova et al., 2015 for further details).

We measured the oxygen isotopic compositions of chemically characterized olivine crystals with a CAMECA ims 1270 E7 at CRPG-CNRS (Nancy, France).  $^{16}\text{O}^-$ ,  $^{17}\text{O}^-$ , and  $^{18}\text{O}^-$  ions produced by a  $\text{Cs}^+$  primary ion beam ( $\sim 15 \mu\text{m}$ ,  $\sim 4 \text{ nA}$ ) were measured in multi-collection mode with two off-axis Faraday cups (FC) for  $^{16,18}\text{O}^-$  and the axial FC for  $^{17}\text{O}^-$ . To remove  $^{16}\text{OH}^-$  interference on the  $^{17}\text{O}^-$  peak and to maximize flatness atop the  $^{16}\text{O}^-$  and  $^{18}\text{O}^-$  peaks, the entrance and exit slits of the central FC were adjusted to obtain mass resolution power of  $\sim 7000$  for  $^{17}\text{O}^-$ . The multicollection FCs were set on exit slit 1 (MRP = 2500). Total measurement times were 240 s (180 s measurement + 60 s pre-sputtering). We used three terrestrial standard materials (San Carlos olivine, magnetite and diopside) to define the instrumental mass fractionation line for the three oxygen isotopes and correct for instrumental mass fractionation for olivine. To obtain good precision on analytical measurements, we analyzed, in order, 4 standards, 8 chondrule olivine crystals and 4 standards. Typical count rates obtained on the San Carlos olivine standards were  $2.5 \times 10^9$  cps for  $^{16}\text{O}$ ,  $1.0 \times 10^6$  cps for  $^{17}\text{O}$ , and  $5.4 \times 10^6$  cps for  $^{18}\text{O}$ . The number of Mg-rich olivines analyzed per chondrule were 45, 45 and 28, with typical uncertainties of 0.2, 0.4 and 0.7‰ for  $\delta^{18}\text{O}$ ,  $\delta^{17}\text{O}$  (measured relative to standard mean ocean water, SMOW, as  $\delta^{17,18}\text{O} = [(^{17,18}\text{O}/^{16}\text{O})_{\text{sample}} / (^{17,18}\text{O}/^{16}\text{O})_{\text{SMOW}} - 1] \times 1000 \text{‰}$ ), and  $\Delta^{17}\text{O} (= \delta^{17}\text{O} - 0.52 \times \delta^{18}\text{O})$ , respectively (Table S1).

## 3. Results

All porphyritic chondrule textural types are present in NWA 5958, i.e., porphyritic olivine-rich (PO), porphyritic olivine-pyroxene (POP), and porphyritic pyroxene-rich (PP); Figs. 1–3). Among the chondrules examined, two PO chondrules (Ch-1 and

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