

# Deciphering recycling processes during solar system evolution from magnesium-rich relict olivine grains in type II chondrules

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#### **Abstract**

Ferromagnesian chondrules present a remarkable dichotomy between reduced (type I) and oxidized (type II) varieties. How these formed, and how they may be related remains contentious. Many type II chondrules, especially in carbonaceous chondrites, contain forsteritic grains in disequilibrium with FeO-rich host olivine grains, which must be relicts of precursor material. In this study, we analyzed the oxygen isotopic composition of magnesian relict and host olivine grains in type II chondrules in CO and CR chondrites. The analyzed relicts are generally more  $^{16}$ O-rich than host grains and plot in the range (isotopic and mineral chemical) of type I chondrules in carbonaceous chondrites. Remarkably, they tend to cluster around the dominant  $\Delta^{17}$ O peaks of the type I chondrules in their host chondrites, viz. -6 % and -2 % for CO and CR, respectively. With the occurrence of relatively intact type I chondrules within some type II chondrules, this corroborates that local type I chondrules were among the precursors of type II chondrules, and that chondrule formation occurred within the accretion reservoir of the eventual chondrites. This supports the nebular brand of chondrule-

forming scenarios. Since not all previous generations of chondrules (or other precursor objects) have been recycled, chondrule formation events must also have been extremely localized. Keywords Carbonaceous chondrites, chondrules, oxygen isotopes, recycling processes, protoplanetary disk 

#### 1- Introduction

The evolution of the circumsolar disk led to the formation of solids through a wide range of processes such as evaporation/condensation (Ebel et al., 2018; Marrocchi et al., 2019a), fusion/crystallization (Connolly & Jones, 2016) and thermal annealing (Han & Brearley, 2015). The resulting dust provided the various building blocks of asteroids and planets, either in the form of refractory inclusions (i.e., CAIs = Calcium Aluminum-rich Inclusions and AOAs = Amoeboid Olivine Aggregates), chondrules and fine-grained matrix. The mineralogical, petrographic, and isotopic features of these early solids thus represent a powerful proxy for deciphering the conditions that prevailed in the circumsolar disk and how they evolved with time.

The mm-sized igneous spheroids called chondrules are the most abundant high-temperature components of both non-carbonaceous (NC) and carbonaceous (C) chondrites, which most likely represent material from the inner and outer solar system, respectively (Kruijer et al., 2017; Schneider et al., 2020). This suggests that the whole circumsolar disk was affected by chondrule production and/or redistribution. The most abundant chondrules are ferromagnesian porphyritic chondrules that can be classified to first order into type I and type II chondrules, depending on their oxidation state (Jones, 2012). Type I chondrules are more reduced than type II chondrules. This translates into high and low Mg# ( $\equiv$  100 × molar ratio Mg/(Mg+Fe)) for type I and type II chondrules, respectively. While the limit between type I and type II is arbitrarily defined as Mg# = 90, there is a hiatus between the two types, with few chondrules between Mg# of 90 and 97 (e.g., Villeneuve et al., 2015). Type II chondrules are predominantly observed in ordinary chondrites but are consistently present in all types of carbonaceous chondrites (Jones, 2012).

Type I chondrules are complex objects whose mineralogy, textures, and chemical and isotopic compositions result from multi-step processes involving precursor recycling, melting event(s) and complex gas-melt interactions (Ebel et al., 2018; Libourel & Portail, 2018; Marrocchi et al., 2018, 2019b; Schneider et al., 2020; Tenner et al., 2018; Villeneuve et al., 2020). The mechanism(s) at their origin remain(s) however controversial: they may correspond to melted aggregates of nebular dust (Hewins et al., 2005; Piralla et al., 2021), e.g. because of bow shocks induced by eccentric planetary embryos (Morris et al., 2012), or byproducts of planetary collisions (Johnson et al., 2015; Libourel et al., 2022; Libourel & Krot, 2007). In addition, type I chondrule formation took place over a long period of time (i.e., 3–4 Ma; (Bollard et al., 2017; Piralla et al., 2023) and could have been repeated, as supported by evidence for distinct generations of chondrules in CR chondrites (Marrocchi et al., 2022). Compared to reduced and volatile-depleted type I chondrules, type II chondrules are characterized by FeO-rich silicates and more chondritic abundances of moderately volatile elements (e.g., Jacquet et al., 2015). The more oxidized conditions of type II chondrule formation could result from either formation in regions with higher dust to gas ratios (Schrader et al., 2013) or lower carbon content in the precursors (Connolly et al., 1994). It has been also proposed that type II chondrules derived from their type I counterparts through nebular oxidation shocks (Villeneuve et al., 2020) or within impact-generated vapor plumes (Libourel et al., 2023).

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Oxygen isotopes may provide more constraints into the formation conditions of both type I and type II chondrules and their potential relationship. First, chondrule isotopic compositions are widely separated from those of AOAs and CAIs (very  $^{16}\text{O}$ -rich, with  $\Delta^{17}\text{O} = \delta^{17}\text{O} - 0.52 \times \delta^{18}\text{O}$  around -25 %; e.g., (Krot et al., 2004). There is also a systematic difference between type I and type II chondrules, especially in carbonaceous chondrites, where the former are  $^{16}\text{O}$ -richer than the latter (Libourel et al., 2023; Schrader et al., 2013;

Tenner et al., 2018; Ushikubo & Kimura, 2020). Likewise, there are some differences depending on the reservoir of origin with type I chondrules in (i) CO–CM–CV chondrites having  $\Delta^{17}$ O ranging from – 7 to – 4 ‰ with a maximum observed near – 6 ‰ (Tenner et al., 2018; Ushikubo and Kimura, 2020) and (ii) CR chondrules that are characterized by  $\Delta^{17}$ O varying from – 6 to –1 ‰ and peaking circa – 2 ‰ (Marrocchi et al., 2022; Schrader et al., 2014; Tenner et al., 2015). The recent characterization of the chemical and isotopic compositions of relict olivine grains in different carbonaceous chondrites suggest that type I chondrules derived (in part) from AOA-like precursors (Marrocchi et al., 2018, 2019b), in agreement with (i) trace element signatures ((Jacquet, 2021; Jacquet & Marrocchi, 2017; Ruzicka et al., 2007) and (ii)  $^{50}$ Ti and  $^{54}$ Cr excesses (Schneider et al., 2020).

<sup>16</sup>O-rich forsteritic olivine grains have been occasionally reported in type II chondrules ((Jones, 1996, 2012; Kunihiro et al., 2004, 2005; Libourel et al., 2023; Schrader et al., 2015, 2018; Wasson & Rubin, 2003; Yurimoto & Wasson, 2002). The similitude of such grains with type I chondrules in the same chondrites have long suggested that these were relicts of type I chondrules, hence highlighting a potential link between type I and type II chondrules. This was also proposed based on the similar large silicon isotopic variations observed within type I chondrules and in Mg-rich olivine relict grains in type II chondrules (Villeneuve et al., 2020). However, no systematic chemical and oxygen isotopic characterization of Mg-rich relict olivine grains in type II chondrules has been performed. In this study, we report a comprehensive survey of the oxygen isotopic compositions of relict-bearing type II chondrules in carbonaceous chondrites, where they are proportionally most abundant, specifically in CO and CR chondrites. We use our data to quantify the conditions of formation of type II chondrules and discuss the implications for the reprocessing of primordial dust in the outer circumsolar disk.

#### 2- Material and Methods

We surveyed all chondrules within (i) four home-made thick sections of CO3 carbonaceous chondrites (Los Vientos 123 [CO3.05], Catalina 008 [CO3.1], El Medano 463 and 465 [CO3.2]) and (ii) two thick sections of CR2 carbonaceous chondrites (Renazzo and Dar al Gani 574, sections no. 719sp3 and 3681sp2, respectively; from the Muséum national d'Histoire naturelle, Paris, France). The petrography and mineralogy of type II chondrules were determined by optical microscopy, backscattered electron (BSE) imaging, and X-ray compositional mapping. For the latter two, we used (i) a Zeiss Evo MA10 equipped with an Oxford X-MAX N 20 SDD energy dispersive spectrometer and operating with a 3 nA electron beam accelerated at 15 kV (Universidad de Atacama, Copiapó, Chile) and (ii) a JEOL JSM-6510 scanning electron microscope equipped with a Genesis EDX detector using a 3 nA electron beam accelerated at 15 kV (CRPG-CNRS, Nancy, France).

Thirteen type II chondrules with and without apparent Mg-rich relicts were selected: 8 in COs and 5 in CRs chondrites. Quantitative analyses of Al, Ti, Ca, Cr, Mn, Ni, Mg, Fe, and Si in all olivine grains large enough to be isotopically characterized by SIMS were performed using a CAMECA SX-Five electron microprobe at the CAMPARIS service (Sorbonne University, Paris, France). The microprobe was calibrated using the following natural and synthetic standards for silicate analyses: diopside (Si, Ca, Mg), orthoclase (Al), MnTiO<sub>3</sub> (Mn, Ti), Cr<sub>2</sub>O<sub>3</sub> (Cr), and Fe<sub>2</sub>O<sub>3</sub> (Fe). A 20 nA focused beam, accelerated to 15 kV potential difference, was used for spot analyses of olivine with 20 s analysis times. Detection limits in silicates were 0.03 wt% for SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, CaO and MgO; 0.04 wt% for MnO and TiO<sub>2</sub>, and

0.07 wt% for NiO, Cr<sub>2</sub>O<sub>3</sub> and FeO. The PAP software was used for matrix corrections (Pouchou and Pichoir, 1984).

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We measured the oxygen isotopic compositions of olivine grains with varied iron contents in type II chondrules using Secondary Ion Mass Spectrometry (SIMS). We used the CAMECA IMS 1270 E7 at the Centre de Recherches Pétrographiques et Géochimiques (CRPG-CNRS, Nancy, France). <sup>16</sup>O<sup>-</sup>, <sup>17</sup>O<sup>-</sup>, and <sup>18</sup>O<sup>-</sup> ions produced by a Cs<sup>+</sup> primary ion beam ( $\sim 10 \, \mu \text{m}$ ,  $\sim 2-2.5 \, \text{nA}$ , rastered over  $5 \times 5 \, \mu \text{m}^2$ ) were measured in multi-collection mode using three Faraday cups (FCs). To remove the interference of <sup>16</sup>OH<sup>-</sup> on the <sup>17</sup>O<sup>-</sup> peak and to maximize the flatness atop the <sup>16</sup>O<sup>-</sup> and <sup>18</sup>O<sup>-</sup> peaks, the entrance and exit slits of the central FC were adjusted to obtain mass resolution power (MRP =  $M/\Delta M$ ) of ~7,000 for  $^{17}O^{-}$ . As an additional safeguard against <sup>16</sup>OH interference, a liquid N<sub>2</sub> trap was used to reduce the pressure in the analysis chamber to  $<1\times10^{-8}$  mbar. The two off-axis multicollection FCs, L'2 and H1, were respectively used to measure <sup>16</sup>O<sup>-</sup> and <sup>18</sup>O<sup>-</sup> and were set on exit slit 1 (MRP = 2,500). We used  $10^{10} \Omega$ ,  $10^{12} \Omega$ , and  $10^{11} \Omega$  resistors for the L'2 ( $^{16}O$ ), central ( $^{17}O$ ), and H1 (<sup>18</sup>O) FCs, respectively (see Bouden et al. 2021 for further details). A normal-incidence electron gun was used for charge compensation. Total measurement times were 300 s, comprising 120 s of pre-sputtering and 180 s of analysis. Two terrestrial standards, San Carlos olivine and JV1 diopside, were used to define the mass-dependent fractionation line. The instrumental mass fractionation (IMF) and matrix effect of chondrule olivine grains was corrected using San Carlos olivine and a set of synthetic standard olivines with Mg# ranging from 36 to 100 (Villeneuve et al., 2019, 2020). To monitor any instrumental drift and to achieve good precision, the San Carlos olivine was analyzed after every 10-15 sample analyses. Typical count rates obtained on San Carlos olivine were  $1.0 \times 10^9$  cps,  $4.0 \times 10^5$  cps, and  $2.1 \times 10^6$  cps for  $^{16}$ O,  $^{17}$ O, and  $^{18}$ O, respectively. All SIMS analytical spots were checked thoroughly by scanning electron microscopy, and any spot near fractures, in the mesostasis, or not completely within olivine grains was excluded from the dataset. Oxygen isotopic compositions are expressed in  $\delta$  units as  $\delta^{17,18}O = [(^{17,18}O/^{16}O)_{sample}/(^{17,18}O/^{16}O)_{V-SMOW} - 1] \times 1000$  % where V-SMOW refers to the Vienna Standard Mean Ocean Water value. Samples related by mass-dependent fractionation to the composition of V-SMOW fall on a line with a slope of 0.52 (defining as the Terrestrial Fractionation Line, TFL; Clayton et al., 1991) whereas mass-independent variations are described by  $\Delta^{17}O = \delta^{17}O - 0.52 \times \delta^{18}O$  (representing vertical deviations from the TFL in a triple oxygen isotope diagram). Typical  $2\sigma$  measurement errors, accounting for internal errors on each measurement and the external reproducibility of the standards, were estimated to be  $\sim 0.4$  % for  $\delta^{18}O$ ,  $\sim 0.7$  % for  $\delta^{17}O$ , and  $\sim 0.7$  % for  $\Delta^{17}O$ .

#### 3- Results

#### 3.1- Petrography and chemistry

We examined all type II chondrules in the five thick sections and selected 13 chondrules with minimum evidence of secondary alteration and olivine grains large enough to perform detailed chemical profiles and isotopic analysis (**Figs. 1–2**). The chondrules are fairly rounded but not, in general, perfectly circular in cross-section, and some may be actually compound (e.g., **Fig.1c,f & Fig. 2c,e**; see also Jacquet 2021). They all display porphyritic texture (except EM463\_Ch05, **Fig. 2f**), with several subhedral to euhedral Fe-rich olivine grains immersed in well-preserved glassy mesostases (**Figs. 1–2**). The maximum apparent diameter of chondrules ranges from 250 to 740 µm and 400 to 750 µm in CO and CR chondrites, respectively. Among the 13 type II chondrules, 11 exhibit Mg-Fe zoning with sharply defined Mg-rich cores and abrupt FeO and/or CaO variation in the chemical profiles

(Figs. 1–2), similar to the relict-bearing chondrules described by Jones et al., (2018). Some relict-bearing chondrules (e.g., LoV123\_Ch02, Fig. 1a) display several forsteritic grains showing Mg diffusion and sharper border inside either larger or smaller Fe-rich olivine grains. The position of relict grains is random, and most of them were found as (i) isolated phenocrysts with Fe-rich rims or (ii) clusters of 3 or (sometimes much) more phenocrysts, where each grain is surrounded by a Fe-rich mantle (Figs. 1–2). Sometimes, the Mg-rich olivine is even part of a more or less intact type I chondrule-like object, with enstatite (stricto sensu, i.e., Fs < 10 mol%), mesostasis and/or metal grains (Fig. 1d & Fig. 2a,b,c), at variance with the contention of Libourel et al. (2023) that enstatite virtually never occurs in type II chondrules. Troilite and chromite are the main opaque and accessory phases in all described FeO-rich chondrules. They are contained in the mesostasis and display sizes up to 90  $\mu$ m, except for CR, where troilite reaches up to 200  $\mu$ m.

The mean chemical composition of relict and host olivine grains among the 13 chondrules is shown in **Table 1**. The Mg# of olivine grains varies widely in the selected chondrules, from 24.6 to 98.3 (**Table S1**), depending on whether the olivine is a host or relict grain. Host olivine grains in CO and CR chondrites display mean Mg# ( $\pm$  standard deviation) of 48.9  $\pm$  11.8 (N= 447) and 51.8  $\pm$  6.1 (N= 22), respectively. Previous values estimated in host olivine show somewhat higher Mg# for CO and CR chondrites ( $\sim$ 66 and 71, respectively; Berlin et al., 2011), but this may hinge on our relict-centered chondrule selection, biasing against large uniformly host (i.e., relict-free) phenocrysts (with less ferroan cores), thus confining host analyses to (more ferroan) overgrowths of large grains or small phenocrysts. As expected, while showing a large range of Fa values (1.8 to 41.8 mol% in CO), relict olivine grains tend to be more forsteritic, with mean Mg# of 85.0  $\pm$  10.6 (N= 189) and 84.3  $\pm$  12.9 (N= 15) for CO and CR chondrites. 70 % of relicts in CR chondrules and 38 % of relicts in CO chondrules (**Table S2**) display Fa values < 10 mol%, that is, in type I compositional

territory (Fig. 2), but the other obviously exited it only because of diffusion of Fe from the host chondrule.

Commonly, Mg-rich relicts in type II chondrules are MnO-poor, and slightly CaO-rich compared to their host olivine grains (**Table 1**). Host olivines display higher mean MnO (0.35 and 0.27 wt%, respectively) and  $Cr_2O_3$  (0.42 vs. 0.25 wt%) in CR than CO chondrites, consistent with earlier data (e.g., Berlin et al., 2011; Schrader et al., 2015).

#### 3.2- Oxygen Isotopes

We performed a total of 88 O-isotopic measurements of relicts and host grains (**Table 2**); 26 and 62 spots were made in CR and CO chondrites, respectively. The olivine O-isotopic composition in CO and CR chondrites ranges respectively from -13.1 to +15.2 % and -11.3 to 8.1 % for  $\delta^{18}$ O; and from -14.2 to 5.8 % and -13.1 to 4.4 %, in the case of  $\delta^{17}$ O.  $\Delta^{17}$ O ranges from -7.7 to +0.7 %. In the three-oxygen diagram (**Fig. 3a**), our data plot along the PCM line proposed by Ushikubo et al. (2012).

Most relict olivine grains are  $^{16}\text{O}$ -rich compared to those in their Fe-rich host phenocrysts (**Figs. 3–4**). However, 36 % (4 over 11) of relict-bearing chondrules show an overlap or similar O-isotopes signature. Only one relict grain in the biggest phenocryst of LoV123\_Ch02 (**Fig. 4a**) shows the opposite relation to its host. The mean  $\Delta^{17}\text{O}$  values of relict grains in CO and CR chondrites are -4.7 and -2.8 %, lower than the mean  $\Delta^{17}\text{O}$  values in type II olivine hosts: -2.6 and -0.5 % for CO and CR chondrites, respectively (**Fig. 5**). Analogous difference of the mean  $\Delta^{17}\text{O}$  values have been reported between relict and host grains in type I chondrules for CR (-5.2 and -3.4 %; Marrocchi et al., 2022). The latest indicate a larger  $\Delta^{17}\text{O}$  difference between relict and host grains from type II chondrules (**Figs. 6–8**). Same was observed by Tenner et al. (2013) for type II chondrules in CO3.0 chondrite

Yamato 81020. In our data set, the isotopic compositions do not show any correlation with size or location within chondrules.

Host grains tend to be isotopically uniform within each chondrule, but there are three apparent exceptions, LoV098-Ch02p, Ren\_Ch01 and DaG574\_Ch01 (**Fig. 4**). In the former two, the exception is restricted to one outlying  $^{16}$ O-rich grain ( $\Delta^{17}$ O = -1.3 and -3.0 ‰, respectively). In the latter, three Fe-rich "host" grains show three distinct  $\Delta^{17}$ O values (-4.7, -3.0 and 0.5 ‰; **Table S2**). Two of them in the latter, and the former two may be isotopic relicts in the sense of Ushikubo et al. (2012) but no longer recognizable in Mg# because of Fe diffusion.

Generally, Mg-rich relicts among CO and CR chondrites overlap the oxygen isotopic composition of relict and host olivine grains in type I chondrules (**Figs 3–4**). In particular, all relicts in CR chondrites plot around -6 ‰ or -2 ‰, i.e. the  $\Delta^{17}$ O values characteristic of the two generations of type I CR chondrules ("type I-CO" and "type I-CR") described by Marrocchi et al. (2022). The most  $^{16}$ O-rich grain found (in chondrule Ren\_Ch03) has a mean  $\Delta^{17}$ O value of  $-7.5 \pm 0.4$  ‰. Yet most relicts (8/10 = 80 %) are in type I-CR territory (i.e.,  $\Delta^{17}$ O > -4 ‰; Marrocchi et al., 2022), as are most type I chondrules in CR. In contrast, type II chondrule forsteritic relicts in CO chondrites are mostly in type I-CO territory (i.e.,  $\Delta^{17}$ O < -4 ‰), as are most type I chondrules there (e.g., Tenner et al., 2013; Libourel et al., 2023).

#### **4- Discussion**

## 4.1- The source of Mg-rich relict grains in type II chondrules

Although type II chondrules have been less studied than their type I counterparts, their formation conditions are no less controversial. They have been historically interpreted as

resulting from closed-system, fractionally crystallized droplets heated at near-liquidus temperature and cooled at  $\sim 1-500$  K/h (see Jones et al., 2018 for a review). The larger size of type II chondrules and their higher concentrations of volatile and moderately volatile elements (i.e., Na, K, Mn, Cr, S, and Fe) compared to type I chondrule were used as arguments for suggesting formation in different nebular (sub)-reservoir than type I chondrules (Hewins & Zanda, 2012; Jacquet et al., 2015). Comprehensive oxygen isotopic observations also suggest the formation of type I and type II chondrules in environments characterized by distinct dust/gas ratios and water ice enhancement, type II being formed in location with dustenrichment factors up to ~4000 above solar (Chaumard et al., 2018, 2021; Hertwig et al., 2018; Tenner et al., 2015). This suggested that type I and type II chondrules formed in two different astrophysical settings, possibly on different sides of the snowline (Tenner et al., 2018). However, the common presence of Mg-rich relict olivine grains within type II chondrules (Figs. 1-2) as well as their chemical and isotopic features (Figs. 3-8) suggest that type II chondrules derive from type I chondrules through oxidation shocks<sup>1</sup> and protracted gas-melt interactions (Ruzicka et al., 2008, Schrader et al., 2015, 2018; Ebel et al., 2018; Villeneuve et al., 2020). A comprehensive experimental approach also showed that these processes can account for several prominent features of type II chondrules, such as chemical zoning in olivine grains and the preservation of Mg-rich olivine relict grains (Villeneuve et al., 2015).

Most of our Mg-rich relict olivine grains in type II chondrules are  $^{16}$ O-rich compared to their Fe-rich host phenocrysts (**Figs 3–4**). Mg-rich relict grains with  $\Delta^{17}$ O values < -15 % were also reported in type II chondrules from CO and CV chondrites (Jones et al., 2002; Libourel et al., 2023; Yurimoto & Wasson, 2002), thus advising a possible genetic link with refractory inclusions (Krot, 2019). However, all our relict grains in CO chondrites display

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<sup>&</sup>lt;sup>1</sup> The expression "oxidation shock" should not be construed to imply a shock wave chondrule-forming scenario.

 $\Delta^{17}$ O values ranging from -7.7 to -0.1 ‰ (n = 8), similar to those reported in the pristine CO3.05 Yamato 81020 (-11.4 ‰ <  $\Delta^{17}$ O < 0.2 ‰, n = 11; Kunihiro et al., 2004; Tenner et al., 2013). Lumping this with previous literature data, this indicates that only ~11 % (3 out of 27) of relicts sample  $^{16}$ O-rich dust similar to refractory inclusions (this study; Yurimoto and Wasson, 2002; Kunihiro et al., 2004; Tenner et al., 2013; Libourel et al., 2023).

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Mg-rich relicts in type II chondrules always plot in the range of O-isotopic composition of host olivines in type I chondrules (Figs. 4–6), and mostly near the dominant mode for type I chondrules in the same chondrites (i.e., near  $\Delta^{17}O = -6$  % in CO and  $\Delta^{17}O =$ -2 % in CR; Figs 4-6). Some outlying Fe-rich "host" grains also do as well (for the CR chondrules Ren\_Ch01 and DaG574\_Ch01, Fig. 4i,l) and may have been of the same nature, but their Mg# was likely overprinted by Fe diffusion during chondrule melting. The mineral chemistry of Mg-rich grains in type II chondrules is also similar to type I chondrule olivine, not only in terms of Fa content, but also in minor element content, e.g. depletion in MnO and enrichment in CaO (Figs 5-8). We also observe varying degrees of assimilation of type I chondrules in type II hosts, from relicts corresponding to whole type I chondrules to more or less identifiable forsterite clusters (Figs 1-2). This suggests that Mg-relicts in type II chondrules originate mainly from Mg-rich type I chondrules, pointing toward a genetic link between both types of chondrules with type II being derived from type I chondrules via oxidation shocks inducing the dissolution of Fe-Ni metal beads and the subsequent crystallization of FeO-rich olivine overgrowths (Ruzicka et al., 2008; Schrader et al., 2015, 2018; Villeneuve et al., 2015, 2020; Libourel et al., 2023).

## 4.2 Implications for chondrule formation

Establishing a genetic relationship between type I and type II chondrules does not in itself provide any information on their environments of formation, as both nebular and planetary conditions may be invoked (e.g., Tenner et al., 2015; Libourel et al., 2023). Libourel et al., (2023) recently proposed that both types of chondrules formed within an isotopically heterogeneous impact-generated vapor plume with a type I-CO-like core region (i.e.,  $\Delta^{17}O < -4$  %) gradually mixing with type I-CR surroundings (i.e.,  $\Delta^{17}O > -4$  %). In such a framework (the impact splash scenario; e.g., Sanders & Scott, 2012; or the jetting scenario; Johnson et al. 2015), type II chondrules derive from first-formed type I chondrules following the drop in the vapor plume temperature (in its outer regions) under unbuffered redox conditions (Libourel et al., 2023). However, several observations weaken this interpretation and, in contrast, favor evolving nebular settings and episodic recycling:

1– Type I chondrules in carbonaceous chondrites commonly display relict grains with chemical (AI-Ti-poor) and isotopic ( $\Delta^{17}O < -15$  ‰) similar to olivine grains present within AOAs (**Fig. 9**; Marrocchi et al., 2018, 2019b; Schneider et al., 2020; Jacquet et al., 2021). In addition, isolated type I chondrules from carbonaceous chondrites show variable <sup>50</sup>Ti and <sup>54</sup>Cr nucleosynthetic anomalies, most of which differing from the host chondrite compositions (Schneider et al., 2020). These isotopic variations suggest that type I CC chondrules were partially formed from precursors isotopically similar to <sup>50</sup>Ti-<sup>54</sup>Cr-rich CAIs and AOAs (Gerber et al., 2017; Schneider et al., 2020: Burkhardt et al., 2023). This thus appears difficult to reconcile with the impact scenario as it would imply that nearly each melt droplet from the impact plume ingested unmelted and unvaporized refractory inclusions wandering in the impact vapor plume.

2– The correlation between Mg# and  $\Delta^{17}$ O reported in the literature (e.g., Tenner et al., 2018 and references therein; Kita et al., 2022) for type I chondrules supports systematically different oxygen fugacities in the type I-CO and type I-CR isotopic reservoirs

(e.g., Schrader et al., 2013; Tenner et al., 2015), or different cooling rates (Jacquet et al., 2021) and hence their likely spatial separation. This correlation was however recently denied (Libourel et al., 2022, 2023) based on data from just two chondrules from CO3.05 chondrite Yamato 81020 (since type I-CR compositions are rare there). In one of them (Ch#35; Libourel et al., 2023), the type I-CR oxygen isotopic compositions are found in ~Fo<sub>99.5</sub> olivine cores with type I-CO overgrowths that we would rather interpret as relicts from a previous generation of chondrules. They may derive from possibly originally larger grains which fossilized higher-temperature, more forsteritic compositions at their (surviving) centers, as observed in isolated olivine grains (Jacquet et al. 2021). Indeed, isolated olivine grains are not restricted to type I-CO compositions, as those of Libourel et al. (2023) are (although virtually all isolated refractory forsterites are type I-CO; Jacquet et al. 2021). They need not represent phenocrysts of lost "megachondrules" with which Libourel et al. (2023) populate the innermost regions of their impact plumes, but plausibly crystals expelled from "normal" molten chondrules and which have grown on all sides by recondensation (Jacquet et al. 2021). Libourel et al. (2022, 2023) do not interpret the isotopically distinct olivine cores as relicts, but rather as the earliest crystallized olivine in the same chondrule-forming event (with isotopically evolving ambient gas). It becomes then difficult to understand why the minor element contents (and in particular the refractory minor elements), and thence their cathodoluminescence properties (Libourel and Portail 2018) abruptly change as the isotopic composition of the gas evolved and the overgrowths crystallized. Libourel et al. (2022, 2023) also suggested that the literature Mg#- $\Delta^{17}$ O correlation is an artifact of excluding putative relicts. However, the said relicts are rare, and mostly <sup>16</sup>O-richer than their host, so including them should not really destroy (although it does denature) the correlation. This may be verified in the Marrocchi et al. (2019) compilation (their Fig. 10 with both relict and host data), bearing in mind that some Kaba (CV3.1; Bonal et al., 2006) type I olivine FeO

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concentrations may have been increased by incipient metamorphism. Consequently, chemistry and isotopic compositions are not decoupled among type I chondrules in carbonaceous chondrites.

3– The Libourel et al. (2023) contention that type I chondrule relicts within type II chondrules do not contain enstatite is erroneous (see results), so at least some type I chondrules must have crystallized their whole mineral assemblage before remelting in a type II chondrule-forming environment. This argues against a monotonous cooling history with oxygen fugacity increasing from type I to type II territory. It must be allowed that enstatite is rare in type II chondrules, when compared to forsterite, which likely marks its easier remelting (because of lower melting temperature).

Taken together, this suggests that type I chondrules are likely nebular products formed through complex processes implying recycling of early condensates, addition of dust and gasmelt interactions (Schneider et al., 2020; Marrocchi et al. 2022). If correct, and considering the genetic link between type I and II chondrules, this would also support a nebular origin for type II chondrules. Since the bulk chemistry of type II chondrules is less devolatilized than type I chondrules (e.g., Jacquet et al., 2015), the latter cannot be the sole precursors of the former. We may envision type I chondrules, whole or fragmented mixed with more chondritic (matrix-like) dust (see **Fig. 10**; not unlike the type I-CO to type I-CR chondrule recycling scenario of Marrocchi et al., 2022) which would have been too fine-grained to survive as relicts. The agglomeratic <sup>16</sup>O-rich objects reported in CR chondrites could represent weakly melted versions thereof (Schrader et al., 2018). The oxygen and silicon isotopic heterogeneities observed within type II chondrules containing Mg-rich olivine relics also imply open-system formation with evolving isotopic compositions being controlled by gasmelt interaction processes, in agreement with their substantially enriched volatile contents relative to type I chondrules.

In terms of astrophysical setting, it appears that the formation of type I chondrules and their recycling into type II chondrules require repetitive heating mechanisms producing different oxygen fugacities. Such processes must be extremely localized for producing type II chondrules without recycling all type I chondrules, implying that chondrites comprise objects affected by a variable number of melting episodes. These spatially-limited processes are also illustrated by our data on CR chondrites where two chondrule populations characterized by different sizes and oxygen isotopic compositions were recently reported (Marrocchi et al., 2022). The larger, <sup>16</sup>O-poor type I-CR chondrules appear to have formed late out from smaller, <sup>16</sup>O-rich type I-CO chondrules that initially populated the CR reservoir (Marrocchi et al., 2022; Tenner et al., 2019). Adequate chondrule-forming scenarios in that respect are those of the "nebular" brand, e.g., bow shocks induced by eccentric planetary embryos (Morris et al., 2012), lightning (Johansen & Okuzumi 2017) or the short-circuit instability (McNally et al. 2013).

#### **5- Concluding remarks**

We have determined the oxygen isotope compositions of Mg-rich relict and host grains in type II chondrules in the carbonaceous chondrites Los Vientos 123 (CO3.05), Catalina 008 (CO3.1), El Médano 463 and 465 (CO3), Renazzo (CR2.4) and Dar al Gani 574 (CR2). Our main results are:

1- Type II chondrules are characterized by large, mass-independent, oxygen isotope variations with olivine O-isotopic composition in CO and CR chondrites ranges respectively from -13.1 to +15.2 ‰ and -11.3 to 8.1 ‰ for  $\delta^{18}$ O; and from -14.2 to 5.8 ‰ and -13.1 to 4.4 ‰, in the case of  $\delta^{17}$ O. These ranges are primarily due to Mg-rich relicts generally  $^{16}$ O-richer than the hosts.

2- The Mg-rich relicts overlap the oxygen isotopic composition of host olivine grains in type I chondrules, with the same dominant peaks shown by their respective host chondrites. All analyzed relicts in CR chondrites plot around -6 % or -2 % for  $\Delta^{17}$ O values similar to the two generations of type I CR chondrules previously identified by Marrocchi et al. (2022).

From this, we drew the following inferences:

- 1- The oxygen isotopic composition of Mg-relicts in type II chondrules supports an origin from Mg-rich type I chondrules. This points toward a genetic link between both types of chondrules with type II being derived from type I chondrules *via* oxidation shocks inducing the dissolution of Fe-Ni metal beads and the subsequent crystallization of FeO-rich olivine overgrowths. The oxygen isotopic heterogeneity observed within type II chondrules containing Mg-rich olivine relict grains also implies a formation of their precursors under open-system conditions.
- 2- Combined with the presence of AOA-like <sup>16</sup>O-rich relict olivine grains and large and variable nucleosynthetic anomalies previously reported in type I chondrules, our data support a nebular origin of both type I and II chondrules.
- 3- As illustrated by the presence of CR type II chondrules with Mg-rich relict grains having  $\Delta^{17}$ O characteristics of the two chondrule generations observed in CR chondrites, our data imply that the nebular process at the origin of chondrules must be extremely localized for producing type II chondrules without recycling all type I chondrules.

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477	Acknowledgments
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483	CRPG contribution #XXXX.
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# Figure caption

Fig 1: Backscattered electron images of relic-bearing type II chondrules from LoV 123 (CO3.05), Médano 465 (CO3) and (e-f) Renazzo (CR2.4). (a) LoV123\_Ch02 is a relict-bearing chondrule with at least ~12 Mg-rich relict grains of various size sharply delineated from their enclosing Fe-rich olivine grains. (b) LoV123\_Ch10 show abundant metal beads in the mesostasis and inside host as well as relict olivines. The central large Mg-rich relict crystal contains three rounded metal beads. (c) Ren\_Ch02 is a lobate chondrule, presumably compound as suggested by the sulfide delineating the protuberance at bottom. The latter has a cluster of small Mg-rich relicts, but a bigger grain appears on the left side of the whole chondrule (possibly in another adhesion, i.e. another component chondrule; Wasson et al., 1995).

(d) LoV123\_Ch07 with a large (~100 μm) enstatite relict, with some olivine chadacrysts and sparse metal beads and an augitic overgrowth. (e) Ren\_Ch03 is a relatively large porphyritic chondrule with the most  $^{16}$ O-rich relict grain at  $\Delta^{17}$ O = -7.5 ‰. (f) The lobate chondrule

Ren\_Ch02 display a cluster of Mg-rich olivine phenocrysts whereas each one is surrounded by thin layer of Fe-rich olivine. Tro = Troilite; Kam = Kamacite; Chr = Chromite; Gyp = gypsum; Fa = Fayalite; Fo = forsterite; Meso = mesostasis.

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Fig. 2: Backscattered electron images of relic-bearing type II chondrules from LoV 123 (CO3.05), Médano 465 (CO3). (a-b) LoV123\_Ch18 displays several Mg-rich olivine crystals sparsely distributed over its area and in the center a metal-bearing IBA chondrule relict intruded by an olivine-microlite rich mesostasis of the host. (c) LoV123\_Ch11 is a large Ferich chondrule (~500 µm) containing a central region with large (>200 µm) partly embayed euhedral olivine phenocrysts, a nonporphyritic right bottom region and two relict-bearing olivine clusters at the top, one with coarse forsterites (near the middle) and another with a olivine-chadacryst-bearing enstatite crystal. Possibly the latter was an independent partly melted droplet which collided with a coarser-grained one. (d) LoV123\_Ch15, fine grained ("agglomeratic"; e.g. Schrader et al. 2018) type II chondrule with a sulfide-rich outer region and a large, elongate type IAB porphyritic chondrule-textured relict (~120 µm), with partly oxidized metal beads. (e) EM465\_Ch04 display a type I POP-textured relict, with (olivine chadacryst-laden) enstatite dominating toward its margin (the top protuberance may be an adhesion merged with the main type I chondrule during its original formation episode) and olivine grains displaying thin Fe-rich margins. Other magnesian relict grains occur sparsely throughout the host. Some porosities are filled by (light gray) gypsum. (f) EM465\_Ch05 shows a cluster of Mg-rich relicts on the right, possibly a late adhesion on the main chondrule, and other sparsely distributed relicts, possibly making a second cluster near the lower left. The porosity is filled by anhedral massive gypsum. Spot analysis in SIMS is indicated as orange and blue circles for relict and host olivines. Tro = Troilite; Kam = Kamacite; Chr = Chromite; Gyp = gypsum; Fa = Fayalite; Fo = forsterite; Meso = mesostasis.

**Fig 3:** (a) Triple oxygen isotope plot of the olivine crystals in the 13 analyzed Fe-rich chondrules. Relict grains show mass-independent variations plotting along the Primary Chondrule Mineral (PCM) line. (b-e)  $\Delta^{17}$ O values vs. (b) Al<sub>2</sub>O<sub>3</sub> (c) CaO, (d) Cr<sub>2</sub>O<sub>3</sub>, (e) MnO and (f) Olivine Fa content (=Fe/[Mg + Fe]×100 mol%) in log scale. The estimated composition of type I and type II chondrules and the Terrestrial Fractionation line (TFL; Clayton et al., 1991) are shown for reference.

**Fig 4:** Oxygen three-isotope diagrams for type II chondrules from (a–h) CO and (i–m) CR chondrites. Reference lines are Terrestrial Fractionation (TF; Clayton et al., 1991), and the Primitive Chondrule Minerals (PCM; Ushikubo et al., 2012). The error bars are smaller than marker size.

**Fig. 5:** Curves show probability density function of  $\Delta^{17}$ O of type I host olivines and type II Mg-relict grains in both CO (a) and CR chondrites (b). Data from this study, Schrader et al. (2013); Tenner et al. (2013, 2015); Fukuda et al., (2022); Kunihiro et al. (2004) and Libourel et al. (2022, 2023).

**Fig 6.** The Mg# -  $\Delta^{17}$ O value of relict and host olivines in type I and type II chondrules from CO (a) and CR (b) chondrites. Fa content (=100 × Fe/[Mg + Fe]) is plotted in log scale revealing a transition and overlapping between olivine grains in type I chondrules and relict grains in type II chondrules for carbonaceous chondrites. The probability density function of relict and host grains in type I and II chondrules are show to the right of each chondrite group. Errors are 2σ. Data of type I chondrules in carbonaceous chondrites are from Schrader et al. (2013); Kunihiro et al. (2004); Tenner et al. (2013); Hertwig et al. (2018); Chaumard et al.

(2018); Marrocchi et al. (2018, 2019, 2022); and Libourel et al. (2022, 2023). The TFL is shown as blue line for reference.

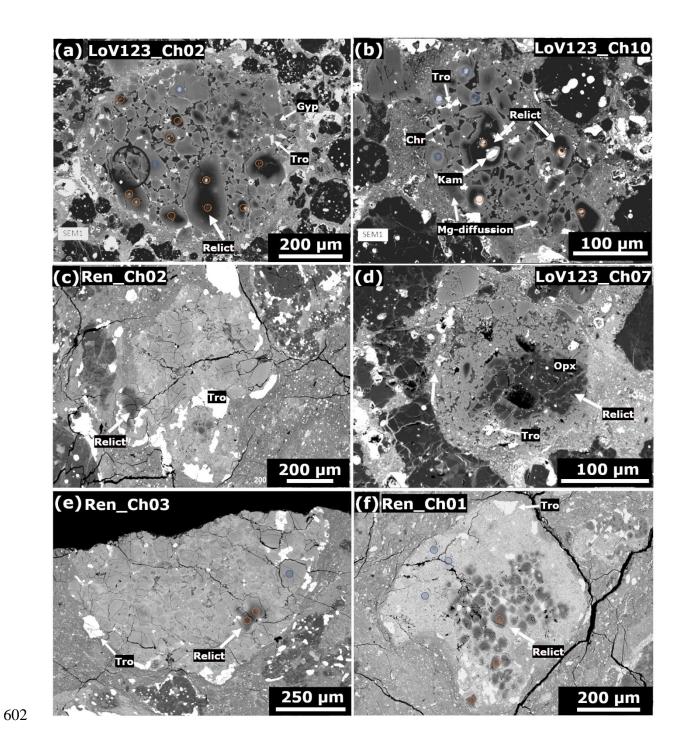
**Fig. 7:** The Δ<sup>17</sup>O value plotted against Al<sub>2</sub>O<sub>3</sub> (a), CaO (b), Cr<sub>2</sub>O<sub>3</sub> (c), and MnO (d) for relict and host olivines in type I and II chondrules from CO, CV, CM and C2-an carbonaceous chondrites. The TFL is shown as blue line for reference. Errors are 2σ. Data of type I and II chondrules of CO, CV, CM and C2-an carbonaceous chondrites from Kunihiro et al. (2004); Tenner et al. (2013); Libourel et al. (2022,2023); Hertwig et al. (2018); Chaumard et al. (2018); Marrocchi et al. (2018, 2019). Data of type II chondrules are show in blue and

skyblue for this study and literature data, respectively.

**Fig. 8:** The  $\Delta^{17}$ O value plotted against Al<sub>2</sub>O<sub>3</sub> (a), CaO (b), Cr<sub>2</sub>O<sub>3</sub> (c), and MnO (d) for relict and host olivines in type I and II chondrules from CR carbonaceous chondrites. The TFL is shown as blue line for reference. Errors are  $2\sigma$ . Data of type I and II chondrules from Schrader et al. (2013) and Marrocchi et al. (2022). Data of type II chondrules are show in green and light green for this study and literature data, respectively.

**Fig. 9:** Schematic representation of formation of type I and II chondrules in CO chondrites. Previously condensed <sup>16</sup>O-rich AOAs mixed with NC-like dust before experienced subsequent melting and gas-melt interactions. This lead to the formation of olivine-rich type I chondrules characterized by <sup>50</sup>Ti-<sup>54</sup>Cr anomalies and the presence of <sup>16</sup>O-rich olivine grains. Depending on the duration of gas-melt interaction, PO, POP or PP type I chondrules are formed. Later heating event under oxidizing conditions induced the formation of type II chondrules. Depending on duration of the oxidation shock, Mg-rich olivine grains can be either conserved or fully dissolved.

Fig. 10: Schematic representation of formation of type I and II chondrules in CR chondrites. Similarly to CO chondrules, small type I CR chondrules result from the melting of a mix of  $^{16}\text{O}$ -rich AOAs and NC-like dust followed by gas-melt interactions. A second generation of large type I chondrules were also formed through the recycling of CI-like dust, inducing the presence of two chondrule generations in CR chondrites. Both type of type I CR chondrules were then recycled into type II chondrules with Mg-rich relict grains having  $\Delta^{17}\text{O}$  characteristics of the two generations of type I chondrule. This implies that the origin of chondrules must be extremely localized for producing type II chondrules without recycling all type I chondrules.



603 Fig. 1

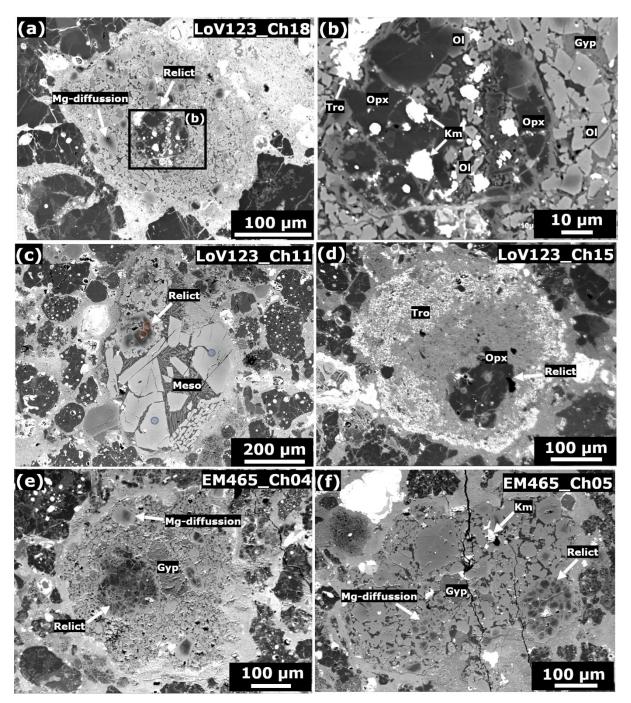


Fig. 2

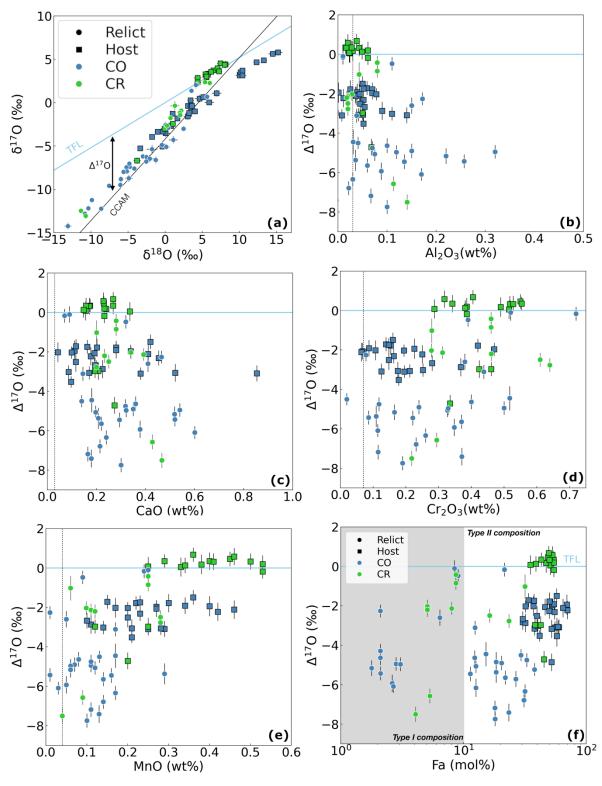
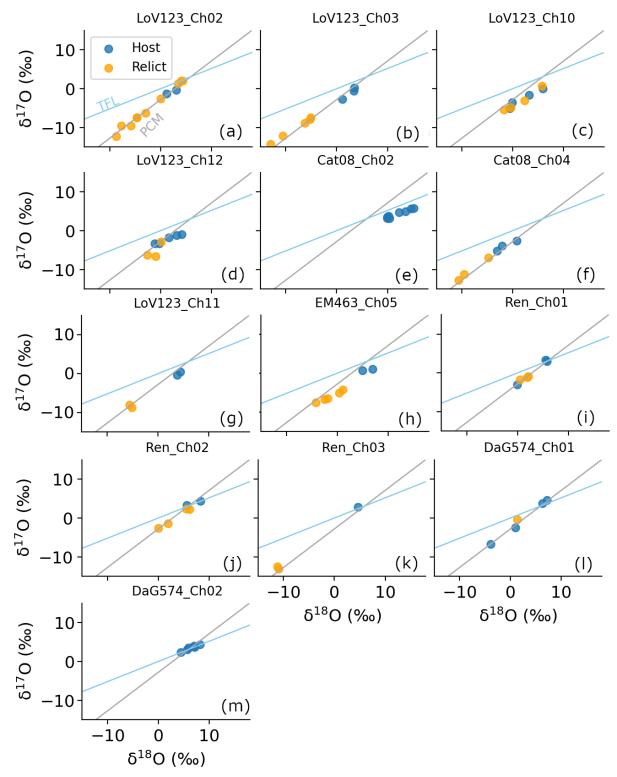
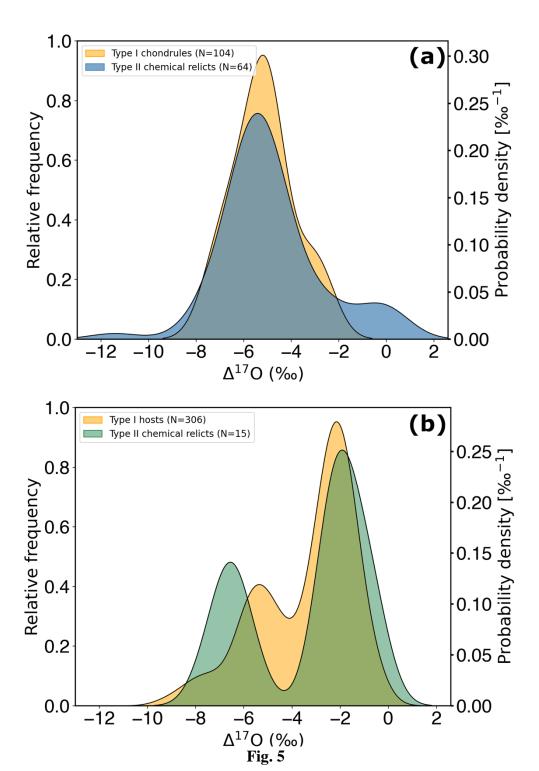


Fig. 3



**Fig. 4** 



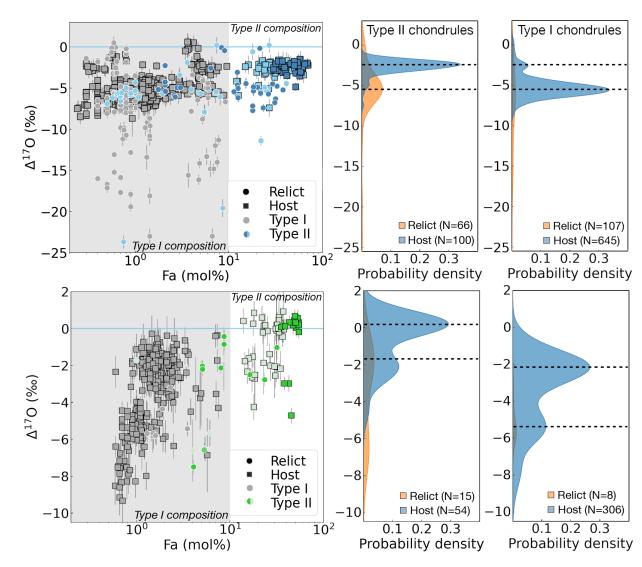


Fig. 6

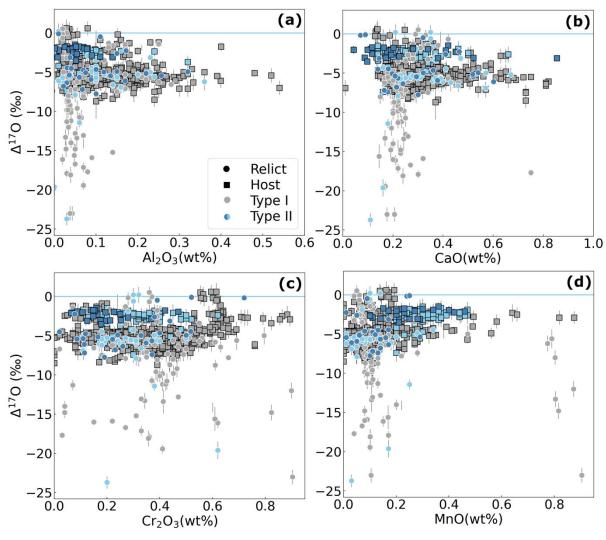


Fig. 7

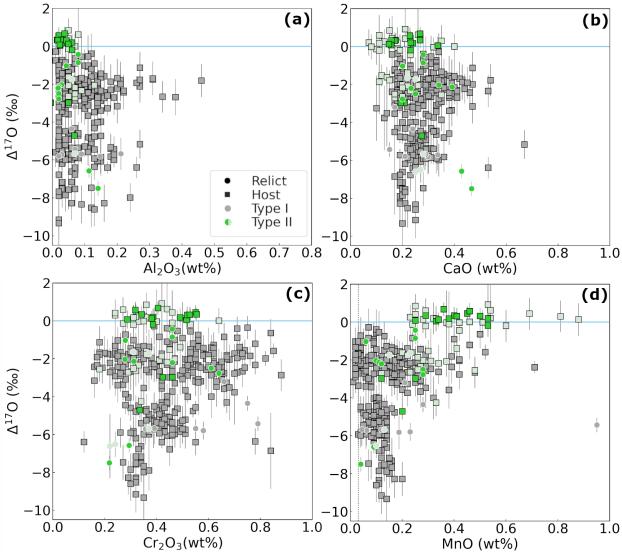


Fig. 8

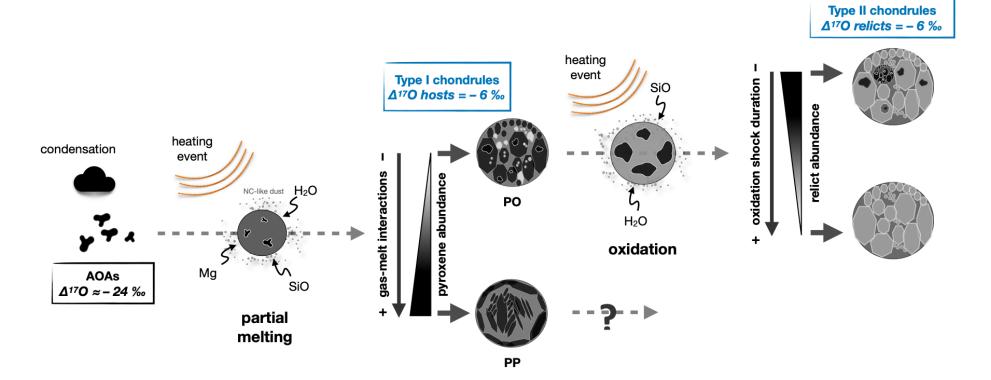
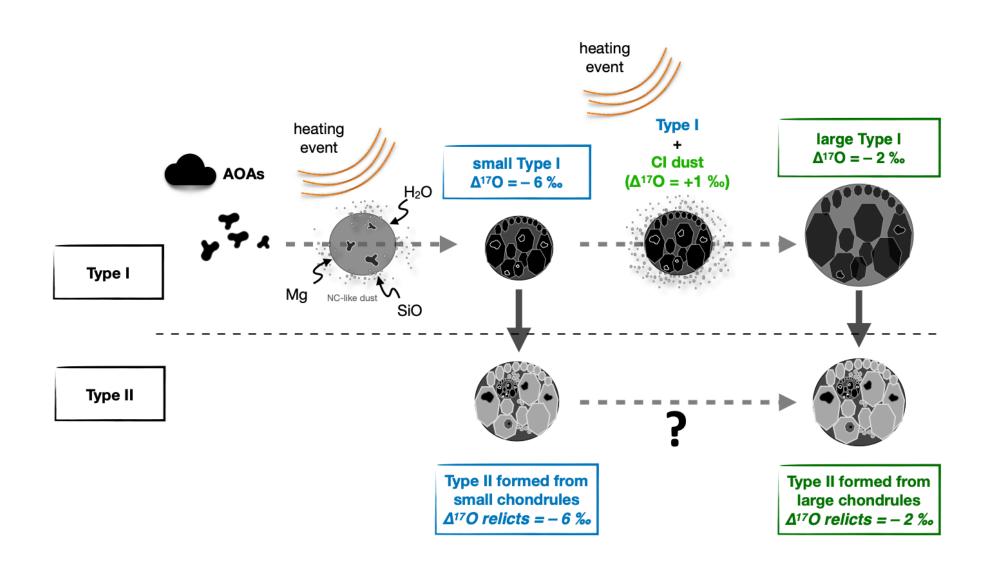


Fig. 9



**Fig. 10** 

Table 1. Mean major and minor composition of 688 spot analysis in host and relict olivine grains among the 13 studied FeO-rich chondrules.

Class	Meteorite	Chondrule	Grain type	N	Mg# (mol%)	MgO	Al <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	CaO	TiO2	Cr2O3	MnO	FeO
СО	LoV123												
		Ch_02	Host	82	51.66	35.52	0.08	37.64	0.35	0.01	0.34	0.23	26.00
			Relict	53	82.78	50.29	0.07	41.34	0.25	0.04	0.37	0.16	8.27
		Ch_12	Host	44	49.40	33.93	0.14	37.03	0.47	0.01	0.30	0.24	27.40
			Relict	20	87.83	52.17	0.09	41.59	0.41	0.03	0.33	0.09	5.92
		Ch_03	Host	72	45.09	31.63	0.15	36.56	0.50	0.02	0.26	0.27	30.04
			Relict	23	72.86	46.04	0.03	39.72	0.34	0.02	0.27	0.14	13.40
		Ch_10	Host	22	42.87	30.00	0.28	36.00	0.45	0.02	0.37	0.23	31.22
			Relict	14	90.20	52.33	0.11	41.74	0.33	0.04	0.49	0.04	4.49
		Ch_11	Host	49	29.54	22.05	0.01	34.34	0.18	b.d.	0.27	0.42	40.98
			Relict	7	84.77	50.41	0.07	40.69	0.23	0.04	0.29	0.14	7.32
	Cat 008												
		Ch_02	Host	108	51.32	34.98	0.07	37.49	0.22	0.01	0.18	0.28	26.40
		Ch_04	Host	50	48.16	33.62	0.06	37.21	0.17	0.01	0.14	0.23	28.57
			Relict	20	84.62	51.20	0.10	41.63	0.37	0.05	0.13	0.08	7.47
	EM463												
		Ch_05	Host	41	52.77	35.85	0.13	37.95	0.17	0.01	0.16	0.25	25.33
			Relict	52	88.64	52.40	0.17	41.67	0.39	0.05	0.14	0.07	5.36
CR	Renazzo												
		Ch_01	Host	4	52.17	35.76	0.01	37.50	0.26	0.02	0.41	0.31	25.52
			Relict	3	93.94	54.93	0.02	41.96	0.32	0.03	0.35	0.11	2.75
		Ch_02	Host	3	53.29	36.07	0.02	37.44	0.19	0.04	0.46	0.37	24.54

		Relict	4	81.57	49.41	0.03	41.02	0.23	0.01	0.55	0.27	8.77
	Ch_03	Host	3	46.16	32.38	0.02	36.86	0.26	0.01	0.38	0.43	29.30
		Relict	3	83.45	49.82	0.08	41.41	0.36	0.06	0.30	0.13	8.28
DaG 574												
	Ch_01	Host	4	55.97	37.99	0.05	38.25	0.23	0.01	0.44	0.24	23.24
		Relict	2	69.09	44.43	0.08	39.39	0.23	0.07	0.39	0.11	15.42
	Ch_02	Host	5	54.17	37.16	0.04	38.11	0.17	b.d.	0.42	0.40	24.58

b.d.:below detection limit

Table 2. Compilation of mean Mg#s and oxygen isotope ratios of 88 spot analysis in the 13 FeO-rich chondrules from the carbonaceous chondrite LoV 123 (CO3.05), Cat 008 (CO3.1), EM 463 (CO3.2), Renazzo (CR2.4), and DaG 574 (CR2).  $\delta^{17}$ O and  $\delta^{18}$ O are relative to VSMOW (Vienna Standard Mean Ocean Water).

Class	Chondrite	Chondrule	Grain type	N analysis	δ <sup>18</sup> Ο (‰)	2σ	δ <sup>17</sup> Ο (‰)	2σ	Δ <sup>17</sup> Ο (‰)	2σ	Mg# (mol%
СО	LoV123										
		Ch02	Relict	10	-2.27	0.31	-4.94	0.33	-3.75	0.37	84.74
			Host	2	2.03	0.33	-0.81	0.29	-1.87	0.33	48.29
		Ch03	Relict	5	-7.96	0.37	-10.12	0.32	-5.98	0.38	69.62
			Host	3	2.56	0.39	-0.97	0.29	-2.30	0.35	43.91
		Ch10	Relict	4	1.48	0.43	-3.14	0.35	-3.91	0.42	94.61
			Host	4	2.17	0.60	-2.53	0.35	-3.66	0.47	45.09
		Ch11	Relict	2	-5.26	0.40	-8.34	0.34	-5.61	0.40	87.39
			Host	2	4.12	0.58	0.06	0.38	-2.08	0.49	30.16
		Ch12	Relict	3	-1.13	0.36	-5.25	0.33	-4.67	0.38	93.97
			Host	5	1.50	0.41	-2.14	0.34	-2.92	0.40	61.91
	Cat008										
		Ch02	Host	8	12.04	0.49	4.31	0.30	-1.95	0.40	53.85
		Ch04	Relict	3	-8.50	0.31	-10.34	0.29	-5.92	0.33	83.31
			Host	3	-1.44	0.32	-3.95	0.27	-3.20	0.32	53.06
	EM463										
		Ch05	Relict	6	-2.04	0.45	-6.37	0.34	-5.31	0.41	89.51
			Host	2	6.02	0.67	0.87	0.36	-2.26	0.50	61.63
CR	Renazzo										
		Ch01	Relict	3	1.61	0.35	-1.28	0.28	-2.12	0.34	93.94
			Host	3	3.75	0.55	1.10	0.35	-0.85	0.45	50.71

	Ch02	Relict	4	3.40	0.36	0.14	0.29	-1.63	0.35	85.73
		Host	2	6.78	0.47	3.85	0.41	0.32	0.43	52.03
	Ch03	Relict	2	-11.02	0.36	-12.76	0.32	-7.03	0.37	95.31
		Host	1	4.36	0.49	2.85	0.36	0.58	0.44	46.87
DaG574										
	Ch01	Relict	1	1.30	0.61	-0.35	0.53	-1.02	0.62	68.32
		Host	4	2.79	0.43	-0.21	0.30	-1.66	0.37	55.97
	Ch02	Host	6	6.41	0.47	3.48	0.34	0.15	0.42	52.74

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