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Gabriel A Pinto, Emmanuel Jacquet, Alexandre Corgne, Felipe Olivares, Johan Villeneuve, et al.. Deciphering recycling processes during solar system evolution from magnesium-rich relict olivine grains in type II chondrules. Geochimica et Cosmochimica Acta, 2024, 364, pp.65 - 78. 10.1016/j.gca.2023.11.012 . hal-04636884

HAL Id: hal-04636884 https://hal.science/hal-04636884

Submitted on 5 Jul2024

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1 2	Deciphering recycling processes during solar system evolution from Mg-rich relict olivine grains in type II chondrules
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17	Abstract
18 19	Ferromagnesian chondrules present a remarkable dichotomy between reduced (type I)
20	and oxidized (type II) varieties. How these formed, and how they may be related remains
21	contentious. Many type II chondrules, especially in carbonaceous chondrites, contain
22	forsteritic grains in disequilibrium with FeO-rich host olivine grains, which must be relicts of
23	precursor material. In this study, we analyzed the oxygen isotopic composition of magnesian
24	relict and host olivine grains in type II chondrules in CO and CR chondrites. The analyzed
25	relicts are generally more ¹⁶ O-rich than host grains and plot in the range (isotopic and mineral
26	chemical) of type I chondrules in carbonaceous chondrites. Remarkably, they tend to cluster
27	around the dominant Δ^{17} O peaks of the type I chondrules in their host chondrites, viz6 ‰
28	and -2 ‰ for CO and CR, respectively. With the occurrence of relatively intact type I
29	chondrules within some type II chondrules, this corroborates that local type I chondrules were
30	among the precursors of type II chondrules, and that chondrule formation occurred within the
31	accretion reservoir of the eventual chondrites. This supports the nebular brand of chondrule-

32	forming scenarios. Since not all previous generations of chondrules (or other precursor
33	objects) have been recycled, chondrule formation events must also have been extremely
34	localized.
35 36	
37	Keywords
38	Carbonaceous chondrites, chondrules, oxygen isotopes, recycling processes, protoplanetary
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70 **1- Introduction**

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73 The evolution of the circumsolar disk led to the formation of solids through a wide 74 range of processes such as evaporation/condensation (Ebel et al., 2018; Marrocchi et al., 75 2019a), fusion/crystallization (Connolly & Jones, 2016) and thermal annealing (Han & 76 Brearley, 2015). The resulting dust provided the various building blocks of asteroids and 77 planets, either in the form of refractory inclusions (i.e., CAIs = Calcium Aluminum-rich 78 Inclusions and AOAs = Amoeboid Olivine Aggregates), chondrules and fine-grained matrix. 79 The mineralogical, petrographic, and isotopic features of these early solids thus represent a 80 powerful proxy for deciphering the conditions that prevailed in the circumsolar disk and how 81 they evolved with time.

82 The mm-sized igneous spheroids called chondrules are the most abundant high-83 temperature components of both non-carbonaceous (NC) and carbonaceous (C) chondrites, 84 which most likely represent material from the inner and outer solar system, respectively 85 (Kruijer et al., 2017; Schneider et al., 2020). This suggests that the whole circumsolar disk 86 was affected by chondrule production and/or redistribution. The most abundant chondrules 87 are ferromagnesian porphyritic chondrules that can be classified to first order into type I and 88 type II chondrules, depending on their oxidation state (Jones, 2012). Type I chondrules are 89 more reduced than type II chondrules. This translates into high and low Mg# (= $100 \times \text{molar}$ 90 ratio Mg/(Mg+Fe)) for type I and type II chondrules, respectively. While the limit between 91 type I and type II is arbitrarily defined as Mg# = 90, there is a hiatus between the two types, 92 with few chondrules between Mg# of 90 and 97 (e.g., Villeneuve et al., 2015). Type II 93 chondrules are predominantly observed in ordinary chondrites but are consistently present in 94 all types of carbonaceous chondrites (Jones, 2012).

95 Type I chondrules are complex objects whose mineralogy, textures, and chemical and isotopic compositions result from multi-step processes involving precursor recycling, melting 96 97 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., 98 99 2020). The mechanism(s) at their origin remain(s) however controversial: they may 100 correspond to melted aggregates of nebular dust (Hewins et al., 2005; Piralla et al., 2021), e.g. 101 because of bow shocks induced by eccentric planetary embryos (Morris et al., 2012), or by-102 products of planetary collisions (Johnson et al., 2015; Libourel et al., 2022; Libourel & Krot, 103 2007). In addition, type I chondrule formation took place over a long period of time (i.e., 3–4 104 Ma; (Bollard et al., 2017; Piralla et al., 2023) and could have been repeated, as supported by 105 evidence for distinct generations of chondrules in CR chondrites (Marrocchi et al., 2022). 106 Compared to reduced and volatile-depleted type I chondrules, type II chondrules are 107 characterized by FeO-rich silicates and more chondritic abundances of moderately volatile 108 elements (e.g., Jacquet et al., 2015). The more oxidized conditions of type II chondrule 109 formation could result from either formation in regions with higher dust to gas ratios 110 (Schrader et al., 2013) or lower carbon content in the precursors (Connolly et al., 1994). It has 111 been also proposed that type II chondrules derived from their type I counterparts through 112 nebular oxidation shocks (Villeneuve et al., 2020) or within impact-generated vapor plumes 113 (Libourel et al., 2023).

114 Oxygen isotopes may provide more constraints into the formation conditions of both 115 type I and type II chondrules and their potential relationship. First, chondrule isotopic 116 compositions are widely separated from those of AOAs and CAIs (very ¹⁶O-rich, with $\Delta^{17}O =$ 117 $\delta^{17}O = 0.52 \times \delta^{18}O$ around -25 ‰; e.g., (Krot et al., 2004). There is also a systematic 118 difference between type I and type II chondrules, especially in carbonaceous chondrites, 119 where the former are ¹⁶O-richer than the latter (Libourel et al., 2023; Schrader et al., 2013;

120 Tenner et al., 2018; Ushikubo & Kimura, 2020). Likewise, there are some differences 121 depending on the reservoir of origin with type I chondrules in (i) CO-CM-CV chondrites having Δ^{17} O ranging from -7 to -4 ‰ with a maximum observed near -6 ‰ (Tenner et al., 122 2018; Ushikubo and Kimura, 2020) and (ii) CR chondrules that are characterized by Δ^{17} O 123 124 varying from -6 to -1 ‰ and peaking circa -2 ‰ (Marrocchi et al., 2022; Schrader et al., 125 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 126 chondrules derived (in part) from AOA-like precursors (Marrocchi et al., 2018, 2019b), in 127 128 agreement with (i) trace element signatures ((Jacquet, 2021; Jacquet & Marrocchi, 2017; Ruzicka et al., 2007) and (ii) ⁵⁰Ti and ⁵⁴Cr excesses (Schneider et al., 2020). 129

¹⁶O-rich forsteritic olivine grains have been occasionally reported in type II chondrules 130 131 ((Jones, 1996, 2012; Kunihiro et al., 2004, 2005; Libourel et al., 2023; Schrader et al., 2015, 132 2018; Wasson & Rubin, 2003; Yurimoto & Wasson, 2002). The similitude of such grains 133 with type I chondrules in the same chondrites have long suggested that these were relicts of 134 type I chondrules, hence highlighting a potential link between type I and type II chondrules. 135 This was also proposed based on the similar large silicon isotopic variations observed within 136 type I chondrules and in Mg-rich olivine relict grains in type II chondrules (Villeneuve et al., 137 2020). However, no systematic chemical and oxygen isotopic characterization of Mg-rich 138 relict olivine grains in type II chondrules has been performed. In this study, we report a 139 comprehensive survey of the oxygen isotopic compositions of relict-bearing type II 140 chondrules in carbonaceous chondrites, where they are proportionally most abundant, 141 specifically in CO and CR chondrites. We use our data to quantify the conditions of formation 142 of type II chondrules and discuss the implications for the reprocessing of primordial dust in 143 the outer circumsolar disk.

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- 147 **2- Material and Methods**
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We surveyed all chondrules within (i) four home-made thick sections of CO3 149 150 carbonaceous chondrites (Los Vientos 123 [CO3.05], Catalina 008 [CO3.1], El Medano 463 151 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 152 153 d'Histoire naturelle, Paris, France). The petrography and mineralogy of type II chondrules 154 were determined by optical microscopy, backscattered electron (BSE) imaging, and X-ray 155 compositional mapping. For the latter two, we used (i) a Zeiss Evo MA10 equipped with an 156 Oxford X-MAX N 20 SDD energy dispersive spectrometer and operating with a 3 nA 157 electron beam accelerated at 15 kV (Universidad de Atacama, Copiapó, Chile) and (ii) a 158 JEOL JSM-6510 scanning electron microscope equipped with a Genesis EDX detector using 159 a 3 nA electron beam accelerated at 15 kV (CRPG-CNRS, Nancy, France).

160 Thirteen type II chondrules with and without apparent Mg-rich relicts were selected: 8 161 in COs and 5 in CRs chondrites. Quantitative analyses of Al, Ti, Ca, Cr, Mn, Ni, Mg, Fe, and 162 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 163 164 University, Paris, France). The microprobe was calibrated using the following natural and synthetic standards for silicate analyses: diopside (Si, Ca, Mg), orthoclase (Al), MnTiO₃ (Mn, 165 166 Ti), Cr₂O₃ (Cr), and Fe₂O₃ (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 167 168 silicates were 0.03 wt% for SiO₂, Al₂O₃, CaO and MgO; 0.04 wt% for MnO and TiO₂, and 169 0.07 wt% for NiO, Cr₂O₃ and FeO. The PAP software was used for matrix corrections
170 (Pouchou and Pichoir, 1984).

We measured the oxygen isotopic compositions of olivine grains with varied iron 171 contents in type II chondrules using Secondary Ion Mass Spectrometry (SIMS). We used the 172 173 CAMECA IMS 1270 E7 at the Centre de Recherches Pétrographiques et Géochimiques (CRPG-CNRS, Nancy, France). ¹⁶O⁻, ¹⁷O⁻, and ¹⁸O⁻ ions produced by a Cs⁺ primary ion 174 beam (~10 μ m, ~2–2.5 nA, rastered over 5 × 5 μ m²) were measured in multi-collection mode 175 using three Faraday cups (FCs). To remove the interference of ¹⁶OH⁻ on the ¹⁷O⁻ peak and to 176 maximize the flatness atop the ${}^{16}O^{-}$ and ${}^{18}O^{-}$ peaks, the entrance and exit slits of the central 177 FC were adjusted to obtain mass resolution power (MRP = $M/\Delta M$) of ~7,000 for ¹⁷O⁻. As an 178 additional safeguard against ¹⁶OH⁻ interference, a liquid N₂ trap was used to reduce the 179 pressure in the analysis chamber to $<1 \times 10^{-8}$ mbar. The two off-axis multicollection FCs, L'2 180 and H1, were respectively used to measure ${}^{16}O^{-}$ and ${}^{18}O^{-}$ and were set on exit slit 1 (MRP = 181 2,500). We used $10^{10} \Omega$, $10^{12} \Omega$, and $10^{11} \Omega$ resistors for the L'2 (¹⁶O), central (¹⁷O), and H1 182 183 (¹⁸O) FCs, respectively (see Bouden et al. 2021 for further details). A normal-incidence 184 electron gun was used for charge compensation. Total measurement times were 300 s, 185 comprising 120 s of pre-sputtering and 180 s of analysis. Two terrestrial standards, San Carlos 186 olivine and JV1 diopside, were used to define the mass-dependent fractionation line. The 187 instrumental mass fractionation (IMF) and matrix effect of chondrule olivine grains was 188 corrected using San Carlos olivine and a set of synthetic standard olivines with Mg# ranging 189 from 36 to 100 (Villeneuve et al., 2019, 2020). To monitor any instrumental drift and to 190 achieve good precision, the San Carlos olivine was analyzed after every 10-15 sample analyses. Typical count rates obtained on San Carlos olivine were 1.0×10^9 cps, 4.0×10^5 cps, 191 and 2.1×10^6 cps for ¹⁶O, ¹⁷O, and ¹⁸O, respectively. All SIMS analytical spots were checked 192 193 thoroughly by scanning electron microscopy, and any spot near fractures, in the mesostasis, or

194 not completely within olivine grains was excluded from the dataset. Oxygen isotopic compositions are expressed in δ units as $\delta^{17,18}O = [(^{17,18}O/^{16}O)_{sample}/(^{17,18}O/^{16}O)_{V-SMOW} - 1] \times$ 195 1000 ‰ where V-SMOW refers to the Vienna Standard Mean Ocean Water value. Samples 196 197 related by mass-dependent fractionation to the composition of V-SMOW fall on a line with a 198 slope of 0.52 (defining as the Terrestrial Fractionation Line, TFL; Clayton et al., 1991) whereas mass-independent variations are described by $\Delta^{17}O = \delta^{17}O - 0.52 \times \delta^{18}O$ 199 (representing vertical deviations from the TFL in a triple oxygen isotope diagram). Typical 2σ 200 201 measurement errors, accounting for internal errors on each measurement and the external reproducibility of the standards, were estimated to be ~0.4 ‰ for δ^{18} O, ~0.7 ‰ for δ^{17} O, and 202 ~0.7 % for Δ^{17} O. 203

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205 **3- Results**

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3.1- Petrography and chemistry

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209 We examined all type II chondrules in the five thick sections and selected 13 210 chondrules with minimum evidence of secondary alteration and olivine grains large enough to 211 perform detailed chemical profiles and isotopic analysis (Figs. 1–2). The chondrules are fairly 212 rounded but not, in general, perfectly circular in cross-section, and some may be actually 213 compound (e.g., Fig.1c,f & Fig. 2c,e; see also Jacquet 2021). They all display porphyritic 214 texture (except EM463_Ch05, Fig. 2f), with several subhedral to euhedral Fe-rich olivine 215 grains immersed in well-preserved glassy mesostases (Figs. 1-2). The maximum apparent 216 diameter of chondrules ranges from 250 to 740 µm and 400 to 750 µm in CO and CR 217 chondrites, respectively. Among the 13 type II chondrules, 11 exhibit Mg-Fe zoning with 218 sharply defined Mg-rich cores and abrupt FeO and/or CaO variation in the chemical profiles 219 (Figs. 1-2), similar to the relict-bearing chondrules described by Jones et al., (2018). Some 220 relict-bearing chondrules (e.g., LoV123_Ch02, Fig. 1a) display several forsteritic grains 221 showing Mg diffusion and sharper border inside either larger or smaller Fe-rich olivine grains. 222 The position of relict grains is random, and most of them were found as (i) isolated 223 phenocrysts with Fe-rich rims or (ii) clusters of 3 or (sometimes much) more phenocrysts, 224 where each grain is surrounded by a Fe-rich mantle (Figs. 1-2). Sometimes, the Mg-rich 225 olivine is even part of a more or less intact type I chondrule-like object, with enstatite (stricto 226 sensu, i.e., Fs < 10 mol%), mesostasis and/or metal grains (Fig. 1d & Fig. 2a,b,c), at variance 227 with the contention of Libourel et al. (2023) that enstatite virtually never occurs in type II 228 chondrules. Troilite and chromite are the main opaque and accessory phases in all described 229 FeO-rich chondrules. They are contained in the mesostasis and display sizes up to 90 μ m, 230 except for CR, where troilite reaches up to 200 µm.

231 The mean chemical composition of relict and host olivine grains among the 13 232 chondrules is shown in Table 1. The Mg# of olivine grains varies widely in the selected 233 chondrules, from 24.6 to 98.3 (Table S1), depending on whether the olivine is a host or relict 234 grain. Host olivine grains in CO and CR chondrites display mean Mg# (± standard deviation) 235 of 48.9 ± 11.8 (N= 447) and 51.8 ± 6.1 (N= 22), respectively. Previous values estimated in 236 host olivine show somewhat higher Mg# for CO and CR chondrites (~66 and 71, respectively; 237 Berlin et al., 2011), but this may hinge on our relict-centered chondrule selection, biasing 238 against large uniformly host (i.e., relict-free) phenocrysts (with less ferroan cores), thus 239 confining host analyses to (more ferroan) overgrowths of large grains or small phenocrysts. 240 As expected, while showing a large range of Fa values (1.8 to 41.8 mol% in CO), relict 241 olivine grains tend to be more forsteritic, with mean Mg# of 85.0 ± 10.6 (N= 189) and $84.3 \pm$ 242 12.9 (N= 15) for CO and CR chondrites. 70 % of relicts in CR chondrules and 38 % of relicts 243 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 thehost 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).

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- **3.2- Oxygen Isotopes**
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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 -11.3to 8.1 ‰ for δ^{18} O; and from -14.2 to 5.8 ‰ and -13.1 to 4.4 ‰, in the case of δ^{17} O. Δ^{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 ¹⁶O-rich compared to those in their Fe-rich host 259 260 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 261 LoV123_Ch02 (Fig. 4a) shows the opposite relation to its host. The mean Δ^{17} O values of 262 relict grains in CO and CR chondrites are -4.7 and -2.8 %, lower than the mean Δ^{17} O values 263 in type II olivine hosts: -2.6 and -0.5 ‰ for CO and CR chondrites, respectively (Fig. 5). 264 Analogous difference of the mean Δ^{17} O values have been reported between relict and host 265 grains in type I chondrules for CR (-5.2 and -3.4 ‰; Marrocchi et al., 2022). The latest 266 indicate a larger Δ^{17} O difference between relict and host grains from type II chondrules (**Figs.** 267 268 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 withsize 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 ¹⁶O-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.

278 Generally, Mg-rich relicts among CO and CR chondrites overlap the oxygen isotopic 279 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 Δ^{17} O values characteristic of the 280 281 two generations of type I CR chondrules ("type I-CO" and "type I-CR") described by Marrocchi et al. (2022). The most ¹⁶O-rich grain found (in chondrule Ren_Ch03) has a mean 282 283 Δ^{17} O value of -7.5 ± 0.4 %. Yet most relicts (8/10 = 80 %) are in type I-CR territory (i.e., Δ^{17} O > -4 ‰; Marrocchi et al., 2022), as are most type I chondrules in CR. In contrast, type II 284 chondrule forsteritic relicts in CO chondrites are mostly in type I-CO territory (i.e., $\Delta^{17}O < -4$ 285 286 ‰), as are most type I chondrules there (e.g., Tenner et al., 2013; Libourel et al., 2023).

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288 **4- Discussion**

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

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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 294 resulting from closed-system, fractionally crystallized droplets heated at near-liquidus 295 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 296 297 (i.e., Na, K, Mn, Cr, S, and Fe) compared to type I chondrule were used as arguments for 298 suggesting formation in different nebular (sub)-reservoir than type I chondrules (Hewins & 299 Zanda, 2012; Jacquet et al., 2015). Comprehensive oxygen isotopic observations also suggest 300 the formation of type I and type II chondrules in environments characterized by distinct 301 dust/gas ratios and water ice enhancement, type II being formed in location with dust-302 enrichment factors up to ~4000 above solar (Chaumard et al., 2018, 2021; Hertwig et al., 303 2018; Tenner et al., 2015). This suggested that type I and type II chondrules formed in two 304 different astrophysical settings, possibly on different sides of the snowline (Tenner et al., 305 2018). However, the common presence of Mg-rich relict olivine grains within type II 306 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 shocks¹ and protracted 307 gas-melt interactions (Ruzicka et al., 2008, Schrader et al., 2015, 2018; Ebel et al., 2018; 308 309 Villeneuve et al., 2020). A comprehensive experimental approach also showed that these 310 processes can account for several prominent features of type II chondrules, such as chemical 311 zoning in olivine grains and the preservation of Mg-rich olivine relict grains (Villeneuve et 312 al., 2015).

Most of our Mg-rich relict olivine grains in type II chondrules are ¹⁶O-rich compared to their Fe-rich host phenocrysts (**Figs 3–4**). Mg-rich relict grains with Δ^{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

¹ The expression "oxidation shock" should not be construed to imply a shock wave chondrule-forming scenario.

 Δ^{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 ‰ < Δ^{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 ¹⁶O-rich dust similar to refractory inclusions (this study; Yurimoto and Wasson, 2002; Kunihiro et al., 2004; Tenner et al., 2013; Libourel et al., 2023).

323 Mg-rich relicts in type II chondrules always plot in the range of O-isotopic 324 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 $\Delta^{17}O =$ 325 326 -2 ‰ in CR; Figs 4-6). Some outlying Fe-rich "host" grains also do as well (for the CR 327 chondrules Ren_Ch01 and DaG574_Ch01, Fig. 4i,l) and may have been of the same nature, 328 but their Mg# was likely overprinted by Fe diffusion during chondrule melting. The mineral 329 chemistry of Mg-rich grains in type II chondrules is also similar to type I chondrule olivine, 330 not only in terms of Fa content, but also in minor element content, e.g. depletion in MnO and 331 enrichment in CaO (Figs 5-8). We also observe varying degrees of assimilation of type I 332 chondrules in type II hosts, from relicts corresponding to whole type I chondrules to more or 333 less identifiable forsterite clusters (Figs 1-2). This suggests that Mg-relicts in type II 334 chondrules originate mainly from Mg-rich type I chondrules, pointing toward a genetic link 335 between both types of chondrules with type II being derived from type I chondrules via 336 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, 337 338 2018; Villeneuve et al., 2015, 2020; Libourel et al., 2023).

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4.2 Implications for chondrule formation

342 Establishing a genetic relationship between type I and type II chondrules does not in 343 itself provide any information on their environments of formation, as both nebular and 344 planetary conditions may be invoked (e.g., Tenner et al., 2015; Libourel et al., 2023). Libourel 345 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., $\Delta^{17}O < -$ 346 4 ‰) gradually mixing with type I-CR surroundings (i.e., $\Delta^{17}O > -4$ ‰). In such a 347 framework (the impact splash scenario; e.g., Sanders & Scott, 2012; or the jetting scenario; 348 349 Johnson et al. 2015), type II chondrules derive from first-formed type I chondrules following 350 the drop in the vapor plume temperature (in its outer regions) under unbuffered redox 351 conditions (Libourel et al., 2023). However, several observations weaken this interpretation 352 and, in contrast, favor evolving nebular settings and episodic recycling:

353 1- Type I chondrules in carbonaceous chondrites commonly display relict grains with chemical (Al-Ti-poor) and isotopic ($\Delta^{17}O < -15$ ‰) similar to olivine grains present within 354 AOAs (Fig. 9; Marrocchi et al., 2018, 2019b; Schneider et al., 2020; Jacquet et al., 2021). In 355 addition, isolated type I chondrules from carbonaceous chondrites show variable ⁵⁰Ti and ⁵⁴Cr 356 357 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 358 partially formed from precursors isotopically similar to ⁵⁰Ti-⁵⁴Cr-rich CAIs and AOAs 359 360 (Gerber et al., 2017; Schneider et al., 2020: Burkhardt et al., 2023). This thus appears difficult 361 to reconcile with the impact scenario as it would imply that nearly each melt droplet from the 362 impact plume ingested unmelted and unvaporized refractory inclusions wandering in the 363 impact vapor plume.

364 2– The correlation between Mg# and Δ^{17} O reported in the literature (e.g., Tenner et 365 al., 2018 and references therein; Kita et al., 2022) for type I chondrules supports 366 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., 367 368 2021) and hence their likely spatial separation. This correlation was however recently denied 369 (Libourel et al., 2022, 2023) based on data from just two chondrules from CO3.05 chondrite 370 Yamato 81020 (since type I-CR compositions are rare there). In one of them (Ch#35; Libourel 371 et al., 2023), the type I-CR oxygen isotopic compositions are found in ~Fo₉₉₅ olivine cores 372 with type I-CO overgrowths that we would rather interpret as relicts from a previous 373 generation of chondrules. They may derive from possibly originally larger grains which 374 fossilized higher-temperature, more forsteritic compositions at their (surviving) centers, as 375 observed in isolated olivine grains (Jacquet et al. 2021). Indeed, isolated olivine grains are not 376 restricted to type I-CO compositions, as those of Libourel et al. (2023) are (although virtually 377 all isolated refractory forsterites are type I-CO; Jacquet et al. 2021). They need not represent 378 phenocrysts of lost "megachondrules" with which Libourel et al. (2023) populate the 379 innermost regions of their impact plumes, but plausibly crystals expelled from "normal" 380 molten chondrules and which have grown on all sides by recondensation (Jacquet et al. 2021). 381 Libourel et al. (2022, 2023) do not interpret the isotopically distinct olivine cores as relicts, 382 but rather as the earliest crystallized olivine in the same chondrule-forming event (with 383 isotopically evolving ambient gas). It becomes then difficult to understand why the minor 384 element contents (and in particular the *refractory* minor elements), and thence their 385 cathodoluminescence properties (Libourel and Portail 2018) abruptly change as the isotopic 386 composition of the gas evolved and the overgrowths crystallized. Libourel et al. (2022, 2023) also suggested that the literature Mg#- Δ^{17} O correlation is an artifact of excluding putative 387 relicts. However, the said relicts are rare, and mostly ¹⁶O-richer than their host, so including 388 389 them should not really destroy (although it does denature) the correlation. This may be 390 verified in the Marrocchi et al. (2019) compilation (their Fig. 10 with both relict and host 391 data), bearing in mind that some Kaba (CV3.1; Bonal et al., 2006) type I olivine FeO 392 concentrations may have been increased by incipient metamorphism. Consequently, chemistry
393 and isotopic compositions are not decoupled among type I chondrules in carbonaceous
394 chondrites.

395 3– The Libourel et al. (2023) contention that type I chondrule relicts within type II 396 chondrules do not contain enstatite is erroneous (see results), so at least some type I 397 chondrules must have crystallized their whole mineral assemblage before remelting in a type 398 II chondrule-forming environment. This argues against a monotonous cooling history with 399 oxygen fugacity increasing from type I to type II territory. It must be allowed that enstatite is 400 rare in type II chondrules, when compared to forsterite, which likely marks its easier 401 remelting (because of lower melting temperature).

402 Taken together, this suggests that type I chondrules are likely nebular products formed 403 through complex processes implying recycling of early condensates, addition of dust and gas-404 melt interactions (Schneider et al., 2020; Marrocchi et al. 2022). If correct, and considering 405 the genetic link between type I and II chondrules, this would also support a nebular origin for 406 type II chondrules. Since the bulk chemistry of type II chondrules is less devolatilized than 407 type I chondrules (e.g., Jacquet et al., 2015), the latter cannot be the sole precursors of the 408 former. We may envision type I chondrules, whole or fragmented mixed with more chondritic 409 (matrix-like) dust (see Fig. 10; not unlike the type I-CO to type I-CR chondrule recycling 410 scenario of Marrocchi et al., 2022) which would have been too fine-grained to survive as relicts. The agglomeratic ¹⁶O-rich objects reported in CR chondrites could represent weakly 411 412 melted versions thereof (Schrader et al., 2018). The oxygen and silicon isotopic 413 heterogeneities observed within type II chondrules containing Mg-rich olivine relics also 414 imply open-system formation with evolving isotopic compositions being controlled by gas-415 melt interaction processes, in agreement with their substantially enriched volatile contents 416 relative to type I chondrules.

417 In terms of astrophysical setting, it appears that the formation of type I chondrules and 418 their recycling into type II chondrules require repetitive heating mechanisms producing 419 different oxygen fugacities. Such processes must be extremely localized for producing type II 420 chondrules without recycling all type I chondrules, implying that chondrites comprise objects 421 affected by a variable number of melting episodes. These spatially-limited processes are also 422 illustrated by our data on CR chondrites where two chondrule populations characterized by 423 different sizes and oxygen isotopic compositions were recently reported (Marrocchi et al., 2022). The larger, ¹⁶O-poor type I-CR chondrules appear to have formed late out from 424 smaller, ¹⁶O-rich type I-CO chondrules that initially populated the CR reservoir (Marrocchi et 425 426 al., 2022; Tenner et al., 2019). Adequate chondrule-forming scenarios in that respect are those 427 of the "nebular" brand, e.g., bow shocks induced by eccentric planetary embryos (Morris et 428 al., 2012), lightning (Johansen & Okuzumi 2017) or the short-circuit instability (McNally et 429 al. 2013).

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431 **5- Concluding remarks**

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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:

437 1- Type II chondrules are characterized by large, mass-independent, oxygen isotope 438 variations with olivine O-isotopic composition in CO and CR chondrites ranges respectively 439 from -13.1 to +15.2 ‰ and -11.3 to 8.1 ‰ for δ^{18} O; and from -14.2 to 5.8 ‰ and -13.1 to 440 4.4 ‰, in the case of δ^{17} O. These ranges are primarily due to Mg-rich relicts generally ¹⁶O-441 richer than the hosts. 442 2- The Mg-rich relicts overlap the oxygen isotopic composition of host olivine grains 443 in type I chondrules, with the same dominant peaks shown by their respective host chondrites. 444 All analyzed relicts in CR chondrites plot around -6 % or -2 % for Δ^{17} O values similar to the 445 two generations of type I CR chondrules previously identified by Marrocchi et al. (2022).

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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.

454 2- Combined with the presence of AOA-like ¹⁶O-rich relict olivine grains and large
455 and variable nucleosynthetic anomalies previously reported in type I chondrules, our data
456 support a nebular origin of both type I and II chondrules.

457 3- As illustrated by the presence of CR type II chondrules with Mg-rich relict grains 458 having Δ^{17} O characteristics of the two chondrule generations observed in CR chondrites, our 459 data imply that the nebular process at the origin of chondrules must be extremely localized for 460 producing type II chondrules without recycling all type I chondrules.

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477	Acknowledgments
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479	Original data from this study are available on the Ordar database:
480	https://doi.org/10.24396/ORDAR-93. Nordine Bouden, Michel Fialin and Nicolas Rividi are
481	thanked for technical assistance. This work was funded by l'Agence Nationale de la
482	Recherche through grant CASSYSS ANR-18-CE31-0010-01 (PI Johan Villeneuve). This is
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 μ m) 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 ¹⁶O-rich relict grain at $\Delta^{17}O = -7.5$ %. (f) The lobate chondrule 517 Ren_Ch02 display a cluster of Mg-rich olivine phenocrysts whereas each one is surrounded
518 by thin layer of Fe-rich olivine. Tro = Troilite; Kam = Kamacite; Chr = Chromite; Gyp =
519 gypsum; Fa = Fayalite; Fo = forsterite; Meso = mesostasis.

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521 Fig. 2: Backscattered electron images of relic-bearing type II chondrules from LoV 123 522 (CO3.05), Médano 465 (CO3). (a-b) LoV123_Ch18 displays several Mg-rich olivine crystals 523 sparsely distributed over its area and in the center a metal-bearing IBA chondrule relict 524 intruded by an olivine-microlite rich mesostasis of the host. (c) LoV123_Ch11 is a large Fe-525 rich chondrule (~500 μ m) containing a central region with large (>200 μ m) partly embayed 526 euhedral olivine phenocrysts, a nonporphyritic right bottom region and two relict-bearing 527 olivine clusters at the top, one with coarse forsterites (near the middle) and another with a 528 olivine-chadacryst-bearing enstatite crystal. Possibly the latter was an independent partly 529 melted droplet which collided with a coarser-grained one. (d) LoV123_Ch15, fine grained 530 ("agglomeratic"; e.g. Schrader et al. 2018) type II chondrule with a sulfide-rich outer region 531 and a large, elongate type IAB porphyritic chondrule-textured relict (~120 µm), with partly 532 oxidized metal beads. (e) EM465_Ch04 display a type I POP-textured relict, with (olivine 533 chadacryst-laden) enstatite dominating toward its margin (the top protuberance may be an 534 adhesion merged with the main type I chondrule during its original formation episode) and 535 olivine grains displaying thin Fe-rich margins. Other magnesian relict grains occur sparsely 536 throughout the host. Some porosities are filled by (light gray) gypsum. (f) EM465_Ch05 537 shows a cluster of Mg-rich relicts on the right, possibly a late adhesion on the main chondrule, 538 and other sparsely distributed relicts, possibly making a second cluster near the lower left . 539 The porosity is filled by anhedral massive gypsum. Spot analysis in SIMS is indicated as 540 orange and blue circles for relict and host olivines. Tro = Troilite; Kam = Kamacite; Chr = Chromite; Gyp = gypsum; Fa = Fayalite; Fo = forsterite; Meso = mesostasis. 541

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) Δ^{17} O values vs. (b) Al₂O₃ (c) CaO, (d) Cr₂O₃, (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.

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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.

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Fig. 5: Curves show probability density function of Δ^{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).

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Fig 6. The Mg# - Δ^{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 2 σ . 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. 567 (2018); Marrocchi et al. (2018, 2019, 2022); and Libourel et al. (2022, 2023). The TFL is
568 shown as blue line for reference.

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Fig. 7: The Δ^{17} O value plotted against Al₂O₃ (a), CaO (b), Cr₂O₃ (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 2 σ . 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 Δ^{17} O value plotted against Al₂O₃ (a), CaO (b), Cr₂O₃ (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 2σ . 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.

582

583 Fig. 9: Schematic representation of formation of type I and II chondrules in CO chondrites. Previously condensed ¹⁶O-rich AOAs mixed with NC-like dust before experienced 584 585 subsequent melting and gas-melt interactions. This lead to the formation of olivine-rich type I chondrules characterized by ⁵⁰Ti-⁵⁴Cr anomalies and the presence of ¹⁶O-rich olivine grains. 586 587 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 588 589 chondrules. Depending on duration of the oxidation shock, Mg-rich olivine grains can be 590 either conserved or fully dissolved.

592 Fig. 10: Schematic representation of formation of type I and II chondrules in CR chondrites. 593 Similarly to CO chondrules, small type I CR chondrules result from the melting of a mix of ¹⁶O-rich AOAs and NC-like dust followed by gas-melt interactions. A second generation of 594 large type I chondrules were also formed through the recycling of CI-like dust, inducing the 595 presence of two chondrule generations in CR chondrites. Both type of type I CR chondrules 596 were then recycled into type II chondrules with Mg-rich relict grains having Δ^{17} O 597 598 characteristics of the two generations of type I chondrule. This implies that the origin of 599 chondrules must be extremely localized for producing type II chondrules without recycling all 600 type I chondrules.



Fig. 1

























Class	Meteorite	Chondrule	Grain type	Ν	Mg# (mol%)	MgO	Al2O3	SiO ₂	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

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

		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

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Class	Chondrite	Chondrule	Grain type	N analysis	δ ¹⁸ Ο (‰)	2σ	δ ¹⁷ Ο (‰)	2σ	Δ ¹⁷ 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		

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). δ^{17} O and δ^{18} O are relative to VSMOW (Vienna Standard Mean Ocean Water).

	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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