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2 **Forging inner-disk Al-rich chondrules by interactions of CAI-like melt**
3 **and ambient gas**

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17

18 ABSTRACT

19 The mechanism of gas-melt interactions and the compositions of precursors are key to
20 understanding the formation of chondrules. To shed light on the two enigmas, we studied the
21 petrography, chemistry, and oxygen isotopes of six Al-rich chondrules (ARCs, five glassy and one
22 plagioclase-bearing) in unequilibrated ordinary chondrites (OCs, petrologic subtype: 3.05). The
23 plagioclase-bearing ARC was also investigated with Al-Mg chronology. Elemental zonation and inter-
24 element correlations in glassy mesostasis of two ARCs indicate the condensation of gaseous Mg,
25 SiO, Fe, and Na onto chondrule melt. The plagioclase-bearing ARC appears to display internal
26 mass-independent oxygen isotope fractionation with $\delta^{18}\text{O}$ increasing following the order of mineral
27 crystallization, suggesting partial oxygen isotope exchange with ambient gas during crystallization.
28 Oxygen isotopes of the six ARCs are distributed along a mixing line of slope = 0.99 ± 0.05 , which
29 intersects with calcium-aluminum-rich inclusions (CAIs), consistent with a small portion of OC type IA
30 chondrules, but deviates from other OC ferromagnesium chondrules (FMCs) towards higher $\delta^{17}\text{O}$,
31 suggesting that OC ARCs and some IA chondrules were established by interactions between CAI-
32 like melts and ^{16}O -poor ambient gas, rather than simply remelting solid mixtures of CAI and FMC
33 materials.

34 All ARCs have unfractionated refractory lithophile element patterns with bulk concentrations
35 ranging from $\sim 7 \times \text{CI}$ to $\sim 15 \times \text{CI}$, indicating $\sim 30\text{-}100\%$ of CAI-like materials in their precursors.
36 Their bulk compositions are linearly evolved toward the Mg: SiO $\sim 3:2$ to $2:1$ (in atomic) apex,
37 consistent with adding gaseous Mg and SiO to the chondrule bulk via gas-melt interactions. The
38 back-calculated compositions of the recycled CAI-like materials closely overlap with pyroxene-
39 anorthite-rich CAIs, suggesting that extensive interactions between the melt of pyroxene-anorthite-
40 rich CAI-like materials and ambient gas could make OC ARCs. The Al-Mg age of the plagioclase-
41 bearing ARC is ~ 2.2 Ma after CAIs, similar to typical OC FMCs, suggesting that the refractory
42 component arrived in the OC reservoirs at the end of the chondrule heating events.

43 **Keywords:** Al-rich chondrules; gas-melt interactions; Ca-Al-rich inclusions; ordinary chondrites.

44

45 **1. Introduction**

46 Chondrules are considered to form in dust-rich environments where interactions between
47 chondrule melts and ambient gas are highly frequent (Cuzzi and Alexander, 2006; Alexander et al.,
48 2008). The gas-melt interactions play a fundamental role in shaping the petrographic, chemical, and
49 isotopic features of chondrules. Petrographically, interactions with gaseous Mg and SiO resulted in
50 the formation of mineralogically-zoned chondrules (low-Ca pyroxene rim with an olivine-rich core;
51 e.g., Tissandier et al., 2002; Friend et al., 2016; Barosch et al., 2019) and near-equilibrium epitaxial
52 growth bands in magnesium-rich olivine (Libourel and Portail, 2018; Marrocchi et al., 2018, 2019;
53 Piralla et al., 2021). Chemically, the condensation of gaseous Mg and SiO onto chondrule melt
54 modified the bulk compositions of initial chondrule melts and contributed to the establishment of
55 major element zonation in chondrule mesostasis (e.g., Libourel et al., 2006; Nagahara et al., 2008).
56 Isotopically, the limited mass-dependent fractionation of isotopes of moderately volatile elements
57 suggests chondrule melt evaporation was suppressed by high partial pressures of these elements in
58 ambient gas and possibly buffered by recondensation processes (e.g., Galy et al., 2000; Tachibana
59 and Huss, 2005; Alexander et al., 2008; Russell et al., 2018; Kadlag et al., 2019, 2021; Villeneuve et
60 al., 2020; Martins et al., 2021). Meanwhile, the common oxygen isotope homogeneity (indicates
61 homogeneous in both $\delta^{18,17}\text{O}$ and $\Delta^{17}\text{O}$; $\Delta^{17}\text{O} = \delta^{17}\text{O} - 0.52 \times \delta^{18}\text{O}$) among minerals and glass in
62 chondrules indicates that oxygen isotope exchanges between chondrule melt and ambient gas was
63 efficient (e.g., Kita et al., 2010; Ushikubo et al., 2012; Tenner et al., 2015; Zhang et al., 2022).

64 However, the mechanism of gas-melt interactions during chondrule formation remain poorly
65 understood, partially due to the subsequent melt crystallization process and asteroidal thermal
66 metasomatism that modified their element distributions and mineral chemistries. In contrast to typical
67 ferromagnesium chondrules (FMCs), Al-rich chondrules (ARCs, bulk $\text{Al}_2\text{O}_3 \geq 10$ wt%) in
68 unequilibrated ordinary chondrites (OCs) typically contain abundant glassy mesostasis in addition to
69 olivine, high-Ca pyroxene, and occasionally, spinel (e.g., Bischoff and Keil, 1984; Ebert and Bischoff,
70 2016). The glassy mesostasis in OC ARCs records primary information about the elemental
71 exchange processes between chondrule melt and ambient gas. Direct evidence for the condensation

72 of gaseous Mg, SiO, and alkali elements is supported by concentric zonation of major elements
73 across glassy mesostasis (Nagahara et al., 2008). Furthermore, the significant Al-rich nature of the
74 chondrule melt recorded internal heterogeneity in the mass-independent fractionation of oxygen
75 isotope indicative of partial exchange with ambient gas (Russell et al., 2000; Jiang et al., 2015; Ebert
76 et al., 2022), different from typical OC FMCs that display oxygen isotope homogeneity (Kita et al.,
77 2010; Piralla et al., 2021; Siron et al., 2021, 2022). Thus, a combined chemical and oxygen isotope
78 study of ARCs in the least metamorphosed OCs would further elucidate the characteristics of gas-
79 melt interactions.

80 After revealing the effect of gas-melt interactions on chondrule bulk compositions, the initial
81 compositions of chondrule melts (precursor) could be interpreted. Major- and trace-element
82 compositions of OC ARCs show enrichments in Al_2O_3 and other refractory lithophile elements (RLEs)
83 that occasionally display group II/III rare earth element (REE) patterns (e.g., MacPherson and Huss,
84 2005; Ebert and Bischoff, 2016), suggesting the existence of refractory calcium-aluminum-rich
85 inclusion (CAI)-like materials among their precursors. This argument is further supported by their
86 oxygen isotope ratios ($\delta^{17, 18}\text{O}$ down to $\sim -15\text{ ‰}$), which are relatively more ^{16}O -enriched than typical
87 FMCs ($\delta^{17, 18}\text{O} > 0\text{ ‰}$) towards those of CAIs ($\delta^{17, 18}\text{O} \sim -45\text{ ‰}$) (Russell et al., 2000; Jiang et al.,
88 2015; Ebert et al., 2022). However, the negative or near-zero $\varepsilon^{50}\text{Ti}$ in OC ARCs argues against this
89 refractory component being a typical CAI ($\varepsilon^{50}\text{Ti}$: +2 to +12); instead, it is more likely derived from a
90 ^{50}Ti excess-free CAI-like component that existed exclusively in the inner disk (Ebert et al., 2018).

91 Here we present petrography, mineral chemistry, bulk major- and trace-element
92 compositions, and oxygen isotopes of six OC ARCs; of which, one was also investigated with Al-Mg
93 chronology. This work differs from previous studies on ARCs which are mainly from metamorphosed
94 type 3.1-4 OCs (Russell et al., 1996, 2000; MacPherson and Huss, 2005; Jiang et al., 2015; Ebert
95 and Bischoff, 2016; Ebert et al., 2022). Instead, our study focuses on ARCs in pristine OCs with
96 petrologic types of 3.05, where their mineral chemistries, elemental distributions, and oxygen and
97 magnesium isotope systems remain largely undisturbed. The results provide critical information on

98 gas-melt interaction mechanism during the formation of OC ARCs and the nature of their refractory
99 precursors.

100 **2. Analytical methods**

101 **2.1 Sample descriptions and electron microprobe analyses**

102 Eight polished sections of six OCs studied by Siron et al. (2021, 2022), i.e., NWA 7731
103 (L3.00), NWA 8276 (L3.00), NWA 8649 (LL3.05), QUE 97008 (L3.05), MET 00452 (L/LL 3.05), and
104 MET 00526 (L/LL 3.05), were surveyed for ARCs. Based on the iron-rich olivine compositions (Cr_2O_3
105 contents; Grossman and Brearley, 2005) and the Fe-Ni metal texture and chemistry (Kimura et al.,
106 2008), these OCs are interpreted to be pristine that experienced minimal asteroidal thermal
107 metamorphism (Agee et al., 2013; Ruzicka et al., 2017; Siron et al., 2022). We note that NWA 7731
108 and NWA 8276 are likely 3.05-3.1 according to Fe-Ni metal texture and chemistry (Siron et al., 2022)
109 rather than 3.00 (Agee et al., 2013; Ruzicka et al., 2017). Thus, we consider them to be similar to the
110 3.05 UOCs. Of the ~3000 chondrules present in these sections, only six were found to be Al-rich:
111 NWA 7731_A1, NWA 8276_A1 & A2, NWA 8649_A1, and MET 00526_A1 & A2.

112 Petrography of the six ARCs was examined using a Hitachi S3400 variable-pressure
113 scanning electron microscope (SEM) at the Geoscience Department, University of Wisconsin-
114 Madison (UW-Madison). Energy dispersive spectroscopy (EDS) X-ray elemental mapping was
115 performed for MET 00526_A2, NWA 8276_A2, and NWA 7731_A1. Major and minor element
116 concentrations of constituent phases of ARCs (spinel, olivine, pyroxene, plagioclase, glass, and
117 microcrystalline mesostasis) were determined using a Cameca SXFive field-emission electron probe
118 microanalyzer (FE-EPMA) at UW-Madison. Analyses were conducted with an accelerating voltage of
119 15 kV, beam current of 10-20 nA, and beam diameter ranging from ~200 nm (focused, for individual
120 phases) to 15 μm (defocused, for mesostasis). Background correction was conducted by linear
121 interpolation between high and low background positions. Counting times were 20 or 10 s on peak
122 and 10 or 5 s on each background position. A time-dependent intensity correction was applied for Na
123 in glass. Interference corrections were applied for interference of V $\text{K}\alpha$ by Ti $\text{K}\beta$ and Cr $\text{K}\alpha$ by V $\text{K}\beta$.

124 Each phase was analyzed with a specific set of elements that were calibrated using standards of
125 high-purity natural or synthetic minerals, oxides, or glasses. The analytical details of beam
126 conditions, detection limits, and calibration standards for each phase are in Table S1. Data
127 acquisition and processing was performed using Probe for EPMA analytical software.

128 2.2 SIMS oxygen three-isotope analyses

129 The oxygen three-isotope ratios of constituent phases in ARCs were determined using the
130 CAMECA IMS-1280 at the WiscSIMS lab of UW-Madison. The measurements were conducted in
131 two sessions using multi-collection Faraday Cup (MCFC) and multi-collection FC and electron
132 multiplier (FC-EM). The analytical condition of the MCFC session is similar to those described in Kita
133 et al. (2010) and Zhang et al. (2022). A focused $^{133}\text{Cs}^+$ primary beam with a size of $\sim 12 \mu\text{m}$ and
134 beam intensity of $\sim 2 \text{nA}$ was used. The typical count rate for $^{16}\text{O}^-$ was $(2-3) \times 10^9 \text{ cps}$ (counts per
135 second). Secondary ions $^{16}\text{O}^-$, $^{17}\text{O}^-$, and $^{18}\text{O}^-$ were detected simultaneously using three FCs with
136 feedback resistors of 10^{10} ohm , 10^{12} ohm , and 10^{11} ohm , respectively. The utilization of a 10^{12} ohm
137 resistor reduced thermal noise for FC and significantly improved analytical precision for $\delta^{17}\text{O}$
138 (Goodrich et al., 2019; Bouden et al., 2021; Zhang et al., 2022). The analytical condition of the FC-
139 EM session is similar to those described in Ushikubo et al. (2012), during which additional analyses
140 on MET 00526_A2 were made. A primary ion beam was focused to $\sim 3 \mu\text{m}$ in size with an intensity of
141 $\sim 20 \text{ pA}$. The typical count rate for $^{16}\text{O}^-$ was $(2-3) \times 10^7 \text{ cps}$. Secondary ion $^{16}\text{O}^-$ was detected using
142 an FC with a resistor of 10^{11} ohm , while $^{17}\text{O}^-$ and $^{18}\text{O}^-$ were detected using EMs. In both sessions,
143 mass resolving power (MRP) was set to ~ 5000 for $^{17}\text{O}^-$, under which the peaks of $^{17}\text{O}^-$ and $^{16}\text{OH}^-$
144 were sufficiently resolved. The count rate of $^{16}\text{OH}^-$ was measured at the end of each measurement,
145 and the contribution of its tailing on $^{17}\text{O}^-$ was corrected using the method described in Heck et al.
146 (2010).

147 Each set of 15-20 measurement of unknown samples was bracketed by eight analyses (four
148 before, four after) on San Carlos olivine (SC-OI), giving spot-to-spot reproducibility (2SD, standard
149 deviation) for $\delta^{18}\text{O}$, $\delta^{17}\text{O}$, and $\Delta^{17}\text{O}$ of $\sim 0.25 \text{\textperthousand}$, $\sim 0.3 \text{\textperthousand}$, $\sim 0.3 \text{\textperthousand}$, respectively, in the MCFC session,

150 and of ~0.5 ‰, ~1.0 ‰, ~1.0 ‰, respectively, in the FC-EM session. The instrumental bias of spinel,
151 olivine, pyroxene, plagioclase, and glass in these ARCs was corrected using reference materials
152 (RM) that generally cover their compositional ranges (listed in Table S4). For glass and high-Ca
153 pyroxene in glassy ARCs, which are significantly richer in Al₂O₃ than the glass and diopside
154 compositions of the RMs used, an additional 0.5 ‰ and 0.25 ‰ uncertainties of instrumental bias
155 corrections were applied empirically to their δ¹⁸O and δ¹⁷O, respectively. Following analysis, the
156 SIMS pits were carefully examined with BSE imaging and EDS measurements using the Hitachi
157 S3400 SEM (supplementary material 1). Data obtained from pits overlapping nearby or subsurface
158 phases and cracks were rejected. If multiple analyses on a chondrule show indistinguishable oxygen
159 isotope ratios, the mean values (“host chondrule”) were calculated (Table S5-6). The uncertainty of
160 mean values was calculated following the reduction scheme described in Hertwig et al. (2018).

161 2.3 SIMS Al-Mg isotope analyses

162 The Al-Mg isotope analyses of major minerals in MET 00526_A2 were performed using the
163 CAMECA IMS-1280 SIMS equipped with a radio-frequency (RF) plasma ion source in two sessions:
164 MCFC for spinel and olivine and FC-EM for plagioclase. The power of the RF source was set to 700
165 W. The analytical conditions for both sessions are similar to those described in Fukuda et al. (2020)
166 and Siron et al. (2021). For both sessions, a ¹⁶O⁻ primary ion beam with an intensity of ~0.5 nA and
167 a size of 6 μm was used.

168 In the MCFC session for spinel and olivine, secondary ions ²⁴Mg⁺, ²⁵Mg⁺, and ²⁶Mg⁺ were
169 counted using FCs with a resistor of 10¹² ohm, and ²⁷Al⁺ was counted simultaneously using an FC
170 with a resistor of 10¹¹ ohm. The typical count rate for ²⁴Mg⁺ was (2-4) × 10⁷ cps. Each analysis took
171 ~10 min, including 100 s of presputtering, ~80 s for secondary ions centering, and 500 s for ion
172 signal integration (10 s × 50 cycles). The instrumental biases of spinel and olivine were corrected
173 with a natural Mg-Al-spinel RM (Mg isotopes unknown, δ²⁶Mg and δ²⁵Mg assumed to be 0 ‰; Kita et
174 al., 2012) and a synthetic forsterite RM (HN-OI) with known Mg isotope ratios (Fukuda et al., 2020),
175 respectively. The relative sensitivity factor (RSF), the SIMS-measured ²⁷Al/²⁴Mg ratio relative to that

176 measured with EPMA, of spinel was also corrected (Kita et al., 2012). Since Al₂O₃ in olivine standard
177 is too low to be accurately determined by EPMA under these conditions, RSF of orthopyroxene
178 standard was determined in the same session and applied to olivine (Ushikubo et al., 2013).

179 In the FC-EM session for plagioclase (0.8-1.0 wt% MgO, as determined by EPMA),
180 secondary ions ²⁴Mg⁺ (5-8 × 10⁵ cps) and ²⁷Al⁺ (5-8 × 10⁷ cps) were counted using two FCs with
181 resistors of 10¹² ohm and 10¹¹ ohm, respectively, while ²⁵Mg⁺ and ²⁶Mg⁺ (~1 × 10⁵) were detected
182 using two EMs. The analysis time was ~32 min, including 120 s of presputtering and FC baseline
183 measurement (for ²⁴Mg⁺), ~80 s for secondary ions centering, and 1600 s for ion signal integration (4
184 s × 400 cycles). To maintain constant EM gains relative to the FC detectors during this long data
185 acquisition period, the high voltage of each EM was adjusted at the 20th and 220th cycles using a
186 Cameca software routine. The gain drift for each EM was monitored and corrected using a second
187 discriminator connected parallel to the first one, as described in supplementary material 2 of Siron et
188 al. (2021). The instrumental biases and RSF of plagioclase was corrected using two anorthitic glass
189 standards with MgO contents of 0.5 wt% and 1.0 wt%, respectively (Kita et al., 2012).

190 The measured ²⁵Mg/²⁴Mg and ²⁶Mg/²⁴Mg ratios were normalized by the absolute Mg isotope
191 ratios (0.12663 and 0.13932, Catanzaro et al. (1966)) and expressed as delta notations $\delta^{25}\text{Mg}$ and
192 $\delta^{26}\text{Mg}$, respectively. The excess $\delta^{26}\text{Mg}^*$, corresponding to the fraction of ²⁶Mg due to the decay of
193 ²⁶Al, was calculated as a function of the instrumental bias-corrected $\delta^{25}\text{Mg}$ and $\delta^{26}\text{Mg}$ values:

$$194 \delta^{26}\text{Mg}^* = [(1 + \frac{\delta^{26}\text{Mg}}{1000}) - (1 + \frac{\delta^{25}\text{Mg}}{1000})^{\frac{1}{\beta}}] \times 1000 = \Delta^{26}\text{Mg} \times (1 + \frac{\delta^{25}\text{Mg}}{1000})^{\frac{1}{\beta}}$$

195 where the $\Delta^{26}\text{Mg}$ is the mass-independent fractionation term and the β is the power of the mass
196 fractionation law that is assumed to be 0.5128 (Davis et al., 2015) for plagioclase, spinel, and olivine.
197 The term $\Delta^{26}\text{Mg}$ is nearly equal to $\delta^{26}\text{Mg}^*$ when the natural mass-dependent fractionation measured
198 as $\delta^{25}\text{Mg}$ in chondrules is small ($\leq 1\text{\textperthousand}$; Ushikubo et al., 2017; Tenner et al., 2019). Typical external
199 reproducibility (2SD) of the raw measured $\delta^{25}\text{Mg}$, $\delta^{26}\text{Mg}$, and $\Delta^{26}\text{Mg}$ for spinel RM were 0.12 †,
200 0.20 †, 0.09 †, for HN-OI were 0.07 †, 0.06 †, 0.15 †, and for anorthitic glass (1 wt% MgO) were
201 0.36 †, 0.30 †, 0.79 †, respectively.

202 The initial $^{26}\text{Al}/^{27}\text{Al}$ ratios ($(^{26}\text{Al}/^{27}\text{Al})_0$) of MET 00526_A2 were obtained from an isochron
203 regression using Isoplot 4.15 (Model 1; Ludwig, 2012). Uncertainties for the reported $(^{26}\text{Al}/^{27}\text{Al})_0$
204 values and relative ages are 95 % confidence intervals. The relative age was calculated as:

205

$$\Delta t (\text{Ma}) = \ln \left[\frac{(\frac{^{26}\text{Al}}{^{27}\text{Al}})_{0, \text{CAI}}}{(\frac{^{26}\text{Al}}{^{27}\text{Al}})_{0, \text{Chondrule}}} \right] \times \frac{0.705}{\ln(2)}$$

206 using the canonical CV CAI $(^{26}\text{Al}/^{27}\text{Al})_0$ of 5.25×10^{-5} (Jacobsen et al., 2008; Larsen et al., 2011) and
207 the half-life of ^{26}Al (0.705 Ma, Nishiizumi, 2004).

208 2.4 LA-ICP-MS trace element analysis

209 After SIMS oxygen and Al-Mg isotope analyses, trace element concentrations of the six
210 ARCs (mainly glassy/microcrystalline mesostasis or a mixture of mesostasis and high-Ca pyroxene)
211 were determined by LA-ICP-MS in the ICP-TIMS lab at UW-Madison. A Teledyne-Photon Machines
212 Analyte femtosecond laser equipped with a HelEx LA cell was operated at a wavelength of 266 nm
213 and a pulse-width of ~ 150 fs. The laser spot size was controlled by an adjustable iris and produced
214 ~ 20 μm diameter craters. During the trace-element analysis, the laser conditions used a pulse
215 energy of ~ 10 mJ on the sample surface, 150 laser shots, and a 5 Hz repetition rate for a total
216 ablation time of 30 s per analysis (pit depth ~ 7 -8 μm on NIST 610). Helium (total flow rate ~ 0.35
217 L/min) was used as the carrier gas to transport ablated particles out of the ablation cell. A mixing cell
218 was used upstream of the plasma to smooth the ablation signal, where additional Ar (1 L/min) and
219 N_2 (5 mL/min) gases were introduced. Laser parameters and flow rates were tuned prior to use by
220 ablating NIST 610. The ablated particles were analyzed on Agilent 8900 QQQ ICP-MS. The ICP-MS
221 was tuned using Agilent Tuning Solution sourced through an Aridus II desolvating nebulizer, and a
222 pulse-to-analog calibration was made using in-house trace element solutions. The oxide production
223 rate, monitored by CeO/Ce during tuning, was $<0.02\text{ }\%$; therefore, no correction was applied. The
224 instrument was operated in single quad mode and data was collected as a time-resolved analysis
225 (TRA) with a total of 29 masses (^{28}Si , ^{29}Si , ^{45}Sc , ^{47}Ti , ^{51}V , ^{52}Cr , ^{55}Mn , ^{63}Cu , ^{66}Zn , ^{85}Rb , ^{88}Sr , ^{89}Y , ^{90}Zr ,

226 ^{137}Ba , ^{139}La , ^{140}Ce , ^{141}Pr , ^{146}Nd , ^{147}Sm , ^{153}Eu , ^{157}Gd , ^{159}Tb , ^{163}Dy , ^{165}Ho , ^{166}Er , ^{169}Tm , ^{172}Yb , ^{175}Lu ,
227 and ^{178}Hf) measured each at an integration time of 0.025 s for a total sampling period of 0.7574 s.

228 TRA data were processed using the “lolite V4” software (Paton et al., 2011). ^{29}Si was used
229 as an internal standard. The NIST SRM 612 (Pearce et al., 1997), USGS reference glass BIR-1G
230 (REE \sim 5-10 \times CI; Jochum et al., 2005) and MPI-DING reference glass GOR 128-G (REE \sim 0.5-9 \times
231 CI; Jochum et al., 2006) were used as running standards to bracket unknown sample analyses.
232 Furthermore, the NIST SRM 610 and USGS reference glasses BCR-2G and BHVO-2G were also
233 analyzed at the beginning of the session. Analytical uncertainties, evaluated by the RSDs (relative
234 standard deviation between measured values and reference values) of the BIR-1G and GOR 128-G
235 (Jochum et al., 2005, 2006), are typically $<10\text{ }\%$. Following analysis, the laser pits were inspected
236 by an optical microscope (supplementary material 1).

237 **3. Results**

238 3.1 Petrography

239 *Plagioclase-bearing ARC*; MET 00526_A2 (ellipsoidal-shaped, 430 \times 250 μm) is the only
240 ARC that contains abundant subhedral-euhedral plagioclase crystals (maximum dimension: \sim 45 μm)
241 in this study. Other phases are spinel, olivine, high-Ca pyroxene, and mesostasis of microcrystalline
242 plagioclase and high-Ca pyroxene (Fig. 1a). Spinel is a subhedral heart-shaped crystal (75 \times 50 μm)
243 located at the upper center of this chondrule. Olivine appears either as tablet crystals (up to 45 μm in
244 length) scattered in the chondrule’s interior or as anhedral crystals decorating its lower right surface
245 (Fig. 1b). High-Ca pyroxene occurs as anhedral crystals between plagioclase laths. Scapolite (6-8 wt%
246 Na_2O , 2-6 wt% CI, \sim 0.7 wt% K_2O , 1-4 wt% FeO , 2-4 wt% CaO , 21-31 wt% Al_2O_3 , 54-60 wt% SiO_2 ,
247 determined by EDS) was identified along the grain boundaries, within the mesostasis, and pooled at
248 the lower center of this chondrule (Fig. 1c). The crystallization sequence of minerals inferred from
249 texture is spinel and olivine followed by plagioclase and high-Ca pyroxene, and then microcrystalline
250 mesostasis.

251 *Glassy ARCs*; The remaining five ARCs of this study are round/ovoid-shaped with a size
252 ranging from $\sim 200 \times 200$ μm to $\sim 1100 \times 700$ μm . They are dominated by Ca-Al-rich glass (NWA
253 8649_A1, NWA 7731_A1, and NWA 8276_A2, Figs. 1d, 2, 3) or slightly devitrified glass of
254 microcrystalline high-Ca pyroxene and plagioclase (MET 00526_A1 and NWA 8276_A1, Fig. 1e-h).
255 Glassy mesostasis is mostly isotropic, apart from that in NWA 8276_A2 (Fig. 2). The right portion of
256 NWA 8276_A2 was replaced by an Mg-Al-Si-Ti-Fe-Ni-bearing hydrated phase (analysis total ~ 70
257 wt%), and its interior has mottled regions with micro-sized pores. Minerals embedded in glassy
258 mesostasis are olivine, high-Ca pyroxene, and, occasionally, spinel. Olivine occurs typically as tablet
259 or skeletal crystals scattered in chondrules' interior or located at chondrule peripheral with long axes
260 parallel or perpendicular to their surfaces. In rare cases, olivine crystals align as parallel bars (NWA
261 8276_A1, Fig. 1g) or anhedral crystals decorating chondrule's surfaces (MET 00526_A1, Fig. 1e).
262 High-Ca pyroxene commonly appears as dendritic crystals, especially in NWA 7731_A1 (~ 37.5
263 vol%). Meanwhile, it also occurs as diamond-shaped crystals with TiO_2 zonation (rim is enriched in
264 TiO_2 , Fig. 1f) and blocky crystals in MET 00526_A1. Spinel was found as a tiny (< 10 μm) crystal at
265 the center of NWA 7731_A1 (Fig. 3c) and coarse-grained (up to 70×35 μm) round crystals
266 occasionally containing melt inclusions in NWA 8276_A1 (Fig. 1h).

267 3.2 Mineral & bulk major element abundances

268 Average major element compositions and modal abundances of constituent phases in the six
269 ARCs and their bulk compositions are listed in Table 1. Individual major element analysis results are
270 in Table S2. Mesostasis are Ca-Al-rich (13-19 wt% CaO , 19-26 wt% Al_2O_3) and MgO-poor (4-11
271 wt%), with minor amounts of Na_2O (typically < 2.0 wt%, up to 3.3 wt%), TiO_2 (typically 0.8-1.2 wt%),
272 Cr_2O_3 (< 0.4 wt%), and FeO (typically < 0.6 wt%, up to 3.6 wt%) and trace (< 0.2 wt%) amounts of
273 V_2O_3 , MnO , and K_2O . The mesostasis of NWA 7731_A1 (high-Ca pyroxene-rich) is richer in SiO_2
274 (50-54 wt% vs. 46-52 wt%) than the others. Furthermore, the hydrated region (Fig. 2a) of NWA
275 8276_A2 is depleted in Na_2O (< 0.1 wt% vs. < 2.8 wt%), CaO (< 1 wt% vs. 16-18 wt%), and Al_2O_3

276 (~10 wt% vs. 20-24 wt%) and enriched in FeO (~4 wt% vs. <0.6 wt%), NiO (~4 wt% vs. <0.1 wt%),
277 and K₂O (~0.2 wt% vs. <0.03 wt%) compared to the interior pristine mesostasis.

278 Olivines grains are mostly forsteritic with Fo >99 and, in rare cases, contain > 1 wt% FeO
279 (Fo₉₇₋₉₉ in MET 00526_A1 and NWA 8276_A2). No chemical zonation of FeO was detected.
280 Concentrations of CaO (0.3-0.7 wt%), Al₂O₃ (0.1-0.7 wt%), Cr₂O₃ (<0.3 wt%), TiO₂ (<0.2 wt%), NiO
281 (<0.5 wt%), and MnO (<0.2 wt%) are minor. CaO in olivines of NWA 8276_A1 is systematically
282 higher than in others (0.5-0.7 wt% vs. 0.3-0.5 wt%). Spinel grains are nearly pure Mg-Al-spinel.
283 Compared to spinels in the glassy ARC NWA 8276_A1, the spinel crystal of plagioclase-bearing
284 ARC MET 00526_A2 is richer in TiO₂ (~0.5 wt% vs. ~0.3 wt%) but poorer in FeO (~0.2 wt% vs. ~0.5
285 wt%), Cr₂O₃ (~0.2 wt% vs. ~0.3 wt%), and V₂O₃ (~0.2 wt% vs. ~0.4 wt%). In addition, plagioclase
286 grains in MET 00526_A2 are anorthitic (An₈₈₋₉₈) with 0.8-1.0 wt% MgO and <1 wt% FeO. They have
287 ≤0.7 mol% (mean value: 0.2) excess structural silica (Si₄O₈) component, as calculated using the
288 algorithm described in Siron et al. (2021).

289 High-Ca pyroxenes in the glassy ARCs contain abundant Al₂O₃ (up to 17 wt%) that does not
290 correlate with their TiO₂ contents; in contrast, in the plagioclase-bearing ARC MET 00526_A2, they
291 have lower Al₂O₃ (3.8-7.0 wt%) that display a good correlation with TiO₂ (Fig. 4), similar to those in
292 ARCs from CV chondrites (Zhang et al., 2020b). Cr₂O₃ (<0.6 wt%), FeO (typically <1.0 wt%), and
293 V₂O₃ (<0.2 wt%) are minor, and Na₂O and MnO are almost absent (<0.1 wt%). The diamond-shaped
294 high-Ca pyroxenes in MET 00526_A1 show normal TiO₂ zonation that increases outward from ~1.4
295 wt% to ~3.0 wt%.

296 Bulk major element compositions of the six ARCs were calculated by combining modal
297 mineral abundances for individual chondrules with the average compositions of their constituent
298 minerals (MacPherson and Huss, 2005). The modal mineral abundances are in mass fraction (wt%),
299 which were converted from area fraction (assumed to be equivalent to the volume fraction) by
300 multiplying the density of individual phases (Zhang et al., 2020b). The six ARCs contain 10-18 wt%
301 CaO, 11-22 wt% Al₂O₃, 11-33 wt% MgO, 44-50 wt% SiO₂, 0.6-1.2 wt% TiO₂, <0.24 wt% Cr₂O₃,
302 <0.07 wt% V₂O₃, <0.06 wt% MnO, and no K₂O. The CaO+Al₂O₃+TiO₂ abundance is lowest in the

303 olivine-rich ARC NWA 8276_A1 (~22 wt%) and goes up to ~32-38 wt% in other ARCs. Bulk Na₂O
304 and FeO contents are typically <1 wt%, except for MET 00526_A2 (~1.5 wt%) which contains
305 abundant scapolite in its microcrystalline mesostasis (analyzed with a defocused electron beam).

306 3.3 Mesostasis & bulk trace element abundances

307 Individual LA-ICP-MS analyses on mesostasis of three glassy ARCs (NWA 8276_A1, NWA
308 7731_A1, and NWA 8649_A1) show nearly uniform and unfractionated RLE (including REEs, Zr, Hf,
309 Sc, Y, Ti, Sr, and Ba) abundances of 10-19 × CI, except for V that exhibits varying degrees of
310 depletions (~0.4-3 × CI). Concentrations of moderately volatile elements (MVEs) Cr (~0.2-0.8 × CI),
311 Mn (~0.1-0.4 × CI), and Rb (typically <0.5 × CI) decrease as their volatility increases (Fig. 5a, Table
312 S3). The trace element patterns of mesostasis in NWA 8276_A2 are very similar to the above three
313 ARCs (Fig. 5b), while concentrations of Rb are highly variable (<0.1-4 × CI). No correlation between
314 trace element abundances and petrographic features, i.e., mottled mesostasis and clean glass, was
315 identified. Blocky high-Ca pyroxene in MET 00526_A1 displays LREE-depletion with negative Eu
316 anomaly and enrichments of Sc, Ti, and V (compatible elements), which are complementary to those
317 in the microcrystalline mesostasis. Similarly, a scapolite-rich region of MET 00526_A2 (plagioclase-
318 bearing) shows significantly depleted (~0.3-3 × CI, except Sr, Ba, and Eu) RLE abundances that are
319 complementary to its microcrystalline mesostasis. But another scapolite-rich region shows
320 indistinguishable trace element abundances from the mesostasis; both display a similar pattern to
321 other glassy ARCs except for the depletion of Sr, Ba, and Eu due to the crystallization of plagioclase.

322 Bulk trace element compositions of the six ARCs were determined by multiplying the modal
323 abundances of mesostasis and/or high-Ca pyroxene with their trace element concentrations
324 determined by LA-ICP-MS (Table 2, Table S3). Additional corrections on bulk V, Cr, Ti, and Mn
325 abundances were made by adding their concentrations in olivine and spinel determined by EPMA,
326 which subtly affects the final totals. As a result, the six ARCs show unfractionated bulk RLEs
327 abundances and volatility-dependent depletions of MVEs (V, Cr, Mn, and Rb). Due to the high

328 abundance of olivine, NWA 8276_A1 ($\text{MgO} + \text{SiO}_2 = 77$ wt%, $\sim 7 \times \text{CI}$) has the lowest bulk
329 concentrations of RLEs compared to the others ($\text{MgO} + \text{SiO}_2 = 60\text{-}67$ wt%, $10\text{-}15 \times \text{CI}$).

330 3.4 Major & trace element distributions in glassy mesostasis

331 Transverse EPMA major element and LA-ICP-MS trace element analyses were conducted
332 across the glassy mesostasis of NWA 8276_A2 (Fig. 2c-d, supplementary material 1). MgO , TiO_2 ,
333 and SiO_2 display chondrule-scale concentric zonation, in which MgO and TiO_2 monotonically
334 decrease and SiO_2 increases from the center to the surface (Fig. 2e). Al_2O_3 content remains almost
335 constant at the upper half of this chondrule (surface “A” to center in Fig. 2c) and shows symmetric
336 zonation (23 wt% \rightarrow 21 wt% \rightarrow 24 wt%) at its lower half (center to surface “B” in Fig. 2c). A slight
337 increase of Al_2O_3 and TiO_2 and a decrease of MgO and SiO_2 were observed near the central olivine
338 (mostly plucked during sample preparation). While exhibiting minor irregularity, the FeO content
339 shows an overall concentric zonation that increases outward from the center to the surface like SiO_2
340 (Fig. 2f). In contrast, CaO and Na_2O contents are complementary to each other without displaying
341 chondrule-scale zonation. Na-rich zones generally bracket Na-poor zones, and a significant amount
342 of Na is concentrated at the interface between the hydrated phase and interior glass (interpreted to
343 be an alteration front) (Figs. 2d, f). On the other hand, transverse LA-ICP-MS analyses found weak
344 symmetric zonation of V, Cr, and Mn in its upper mesostasis (surface A to center), while their profiles
345 become irregular towards the lower surface B (Fig. S1). The Ti zonation profile determined by LA-
346 ICP-MS is broadly similar to that determined by EPMA (Figs. 2e, S1). No chemical zonation of other
347 RLEs (including REEs) were detected.

348 Transverse EPMA major element analyses were also performed for the mesostasis of NWA
349 7731_A1. Unlike NWA 8276_A2, its mesostasis displays localized symmetric zonation of MgO , SiO_2 ,
350 Al_2O_3 , and, to a weaker extent, TiO_2 (divided as segments I, II, III, and IV in Fig. 3e). The zonation
351 patterns of segments I, II, and IV that are between high-Ca pyroxene branches are similar to the
352 chondrule-scale zonation patterns observed in NWA 8276_A2, while segment III that crosses a
353 coarse high-Ca pyroxene branch is opposite. In contrast, the FeO and Na_2O contents exhibit

354 concentric zonation across the entire chondrule (Fig. 3f). The peripheral \sim 100 μm region and several
355 interior patches of this chondrule are richer in Na_2O , as illuminated by its X-ray elemental map of Na
356 (Fig. 3d). The Na_2O and CaO abundances are in general complementary, with the latter showing
357 larger variability due to high-Ca pyroxene crystallization.

358 3.4 Oxygen isotopes

359 A total of 43 analyses were performed on the six ARCs using a 12 μm primary beam (MCFC
360 mode), and additional eight analyses were conducted on MET 00526_A2 using a 3 μm primary
361 beam (FC-EM mode). Data from two analyses were rejected due to mixing with the adjacent
362 chondrite matrix or another phase beneath (supplementary material 1). The results of oxygen
363 isotope analyses on two ARCs (MET 00526_A2 and NWA 8649_A1) are variable, while others are
364 homogeneous (Fig. 6, Table 3, S5-S6). The plagioclase-bearing ARC MET 00526_A2 displays the
365 largest oxygen isotope variability, with $\delta^{18}\text{O}$ increasing in the order of spinel (\sim –10 ‰), olivine (\sim
366 9 ‰), high-Ca pyroxene (\sim –8 ‰), and plagioclase (–8 ‰ to –5 ‰), while $\Delta^{17}\text{O}$ increases from –5 ‰
367 to –3.5 ‰. Similar mass-independent fractionation of oxygen isotopes is found in the glassy ARC
368 NWA 8649_A1, which shows ^{16}O -enrichment at the center ($\delta^{18}\text{O}$: –5 ‰; $\Delta^{17}\text{O}$: –4 ‰) compared to
369 the other analyses towards the edge ($\delta^{18}\text{O}$: –1 ‰; $\Delta^{17}\text{O}$: –1 ‰). The remaining four glassy
370 chondrules display internal oxygen isotope homogeneity with $\delta^{18}\text{O}$ and $\Delta^{17}\text{O}$ ranging from –9 ‰ to –
371 1 ‰ and –4 ‰ to –1 ‰, respectively. Two analyses on the mottled region of glassy mesostasis in
372 NWA 8276_A2 show $\delta^{18}\text{O}$ and $\Delta^{17}\text{O}$ of \sim +5 ‰ and \sim +1 ‰, respectively, significantly more positive
373 than coexisting olivine and clean glass (Table 3). All analyses are plotted above the primitive
374 chondrule mineral (PCM) and on/below the terrestrial fractionation (TF) line (Fig. 6).

375 3.5 Al-Mg isotopes

376 A total of 15 Al-Mg isotope analyses on olivine ($N = 2$), spinel ($N = 3$), and plagioclase ($N =$
377 10, three were rejected) from MET 00526_A2 were conducted (Table 4, Table S7-S9). The 12 valid
378 analyses define an isochron ($\text{MSWD} = 1.6$) with an inferred $(^{26}\text{Al}/^{27}\text{Al})_0$ of $(6.3 \pm 1.2) \times 10^{-6}$ and a

379 $\delta^{26}\text{Mg}^*_0$ of $-0.08 \pm 0.07 \text{‰}$ (Fig. 7). Slightly negative initial $\delta^{26}\text{Mg}^*_0$ is caused by the spinel analyses
380 that do not show ^{26}Mg excess ($\sim 0 \text{‰}$) beyond the analytical uncertainty (2SE) of 0.15‰ . The
381 regression line excluding spinel show an inferred $(^{26}\text{Al}/^{27}\text{Al})_0$ of $(6.2 \pm 1.2) \times 10^{-6}$ and a $\delta^{26}\text{Mg}^*_0$ of
382 $0.02 \pm 0.10 \text{‰}$ (MSWD = 1.0), where spinel data overlap within their analytical uncertainties. The
383 isochron using all data corresponds to a formation age of $2.15 \pm 0.22/0.18 \text{ Ma}$ after CAI, under the
384 assumption of a homogeneous distribution of ^{26}Al with a canonical value of 5.25×10^{-5} in the early
385 solar system (Jacobsen et al., 2008; Larsen et al., 2011).

386 4. Discussion

387 4.1 Classification of OC ARCs

388 Petrologic classification of OC ARCs is challenging because most ($\sim 90 \text{ \%}$) are dominated by
389 glassy mesostasis indicative of rapid cooling that does not follow the phase equilibria. Based on their
390 phenocryst types, Krot and Rubin (1994) divided glassy ARCs as glass/cryptocrystalline (90-99 vol%
391 isotropic glass with skeletal crystals of olivine and high-Ca pyroxene), barred-olivine (BO),
392 porphyritic olivine (PO), porphyritic pyroxene (PP), and porphyritic olivine-pyroxene (POP), in
393 resemblance to the classification scheme of FMCs. The OC ARCs studied by Ebert and Bischoff
394 (2016) and Ebert et al. (2022) are all glassy and were grouped as BO, PO, POP, and PP. Following
395 this classification, NWA 8276_A2 of this study is PO, NWA 8276_A1 is BO, and MET 00526_A1 is
396 POP. NWA 7731_A1 and NWA 8649_A1 are better classified as nonporphyritic because their high-
397 Ca pyroxene is dendritic and significant Al-rich, comparable to the metastable high-Ca pyroxene in
398 the run products of a CMAS melt (12.2 wt% CaO, 28.5 wt% MgO, 12 wt% Al_2O_3 , and 47.3 wt% SiO_2)
399 that crystallized under high degree of undercooling (olivine is the liquidus phase; Faure and
400 Tissandier, 2014). Thus, the dendritic high-Ca pyroxene is a metastable phase rather than
401 phenocrysts. The wide occurrence of skeletal olivine in these glassy ARCs also indicates a high
402 degree of undercooling (Faure et al., 2003). Furthermore, the higher portion of glassy chondrules
403 among OC ARCs compared to type I FMCs could be explained by their lower liquidus temperatures
404 ($\sim 1400\text{--}1600 \text{ }^\circ\text{C}$ vs. $\sim 1600\text{--}1800 \text{ }^\circ\text{C}$) (Hewins and Radomsky, 1990), where undercooling of

405 chondrule melts can be more easily achieved under similar cooling environments. For the barred-
406 olivine texture in NWA 8276_A1, it implies either a fast-cooling (100-7200 K/h; Hewins and
407 Radomsky, 1990; Lofgren and Lanier, 1990; Tsuchiyama et al., 2004) followed by multiple reheating
408 steps or a slow-cooling of chondrule melt (<10 K/h; Faure et al., 2022).

409 The less common (~10 %) non-glassy ARCs are composed of well-crystallized plagioclase
410 and high-Ca pyroxene with/without spinel, olivine, and low-Ca pyroxene. MacPherson and Huss
411 (2005) studied eight non-glassy ARCs and divided them into two groups, i.e., porphyritic olivine (PO,
412 N = 6) and porphyritic plagioclase (N = 2), basing on their bulk compositions and phase equilibria.
413 Among the PO ARCs, three (Chainpur 1251-16-2, Chainpur 5674-3b-1, and Krymka 1729-9-1) have
414 only 3-6 wt% bulk Al₂O₃ and should not be considered as ARCs. The remaining three have abundant
415 olivine (Semarkona 4128-3-2 and Chainpur 1251-14-1, 38-46 wt%) or both olivine and low-Ca
416 pyroxene (Quinyambie 6076-5-2, 30 wt% olivine, 37 wt% low-Ca pyroxene) phenocrysts, which are
417 better named as PO and POP ARCs, respectively. For the two porphyritic plagioclase ARCs
418 (Chainpur 5674-2-1 and Chainpur 1251-14-2), they are dominated by plagioclase laths and anhedral
419 high-Ca pyroxene, with much lower olivine abundances than PO ARCs (0-15 wt% vs. 38-46 wt%).
420 MET 00526_A2 of this study has mineral assemblage (spinel, olivine, plagioclase, and high-Ca
421 pyroxene) almost identical to the two PO ARCs, but with a lower olivine abundance (~20 wt% olivine)
422 that transits to porphyritic plagioclase ARCs. Thus, we conclude that the glassy and non-glassy
423 ARCs share the same phenocryst types, and the key difference is their mesostasis types (glass vs.
424 crystallized plagioclase + high Ca pyroxene), which primarily reflects their differences in cooling rate
425 below the glass transition temperature (T_g, the transition from a viscous state to a brittle state)
426 (Tronche et al., 2007; Wick and Jones, 2012) as well as the degree of undercooling as discussed
427 above. For simplicity, we classify OC ARCs as glassy and plagioclase-bearing subtypes.

428 **4. 2 Gas-melt interactions**

429 It is widely accepted that chondrule formation occurred in open systems where interactions
430 between chondrule melt and ambient gas have major effects on the petrologic, chemical, and

431 isotopic characteristics of the final products. Direct petrologic evidence for gas-melt interactions
432 during chondrule formation include (i) mineralogically-zoned chondrules common in carbonaceous
433 and ordinary chondrites, which have been reproduced by exposing chondrule-like melt to a gas of
434 high SiO partial pressure in isothermal condensation experiments, indicating that the peripheral low-
435 Ca pyroxene layer could have been formed by reactions between SiO gas and olivine crystals
436 (Tissandier et al., 2002; Friend et al., 2016; Barosch et al., 2019); (ii) the near-equilibrium epitaxial
437 growth of magnesium-rich olivine revealed by high-resolution CL images, suggesting that the
438 condensation of gaseous Mg and SiO onto the chondrule melt supplied the growth of olivine crystals
439 (Libourel and Portail, 2018; Marrocchi et al., 2018, 2019; Piralla et al., 2021); and (iii) concentric
440 chemical zonation (MgO, SiO₂, Al₂O₃, TiO₂, etc.) across chondrule mesostasis, implying
441 condensation of Mg, Si, Fe, and alkali elements during chondrule formation (Nagahara et al., 2008).

442 4.2.1 Major & trace element evidence for gas-melt interactions

443 Concentric chemical zonation of major elements across the glassy mesostasis was observed
444 in NWA 8276_A2 of this study, with patterns broadly similar to those in the glassy ARC CH5 from
445 Semarkona (Nagahara et al., 2008). Since olivine crystals are mostly located at the chondrule
446 periphery, the crystallization of olivine would consume MgO, SiO₂, and FeO and enrich SiO₂ (bulk
447 melt is richer in SiO₂ than olivine, ~47 wt% vs. ~43 wt%) and other incompatible elements (Al, Ti, Ca,
448 V, Cr, Mn, Na, and K) in nearby melt (Kennedy et al., 1993; Libourel, 1999), resulting in monotonic
449 decreases in MgO and FeO and increases of SiO₂, Al₂O₃, TiO₂, and CaO from the chondrule center
450 to the surface. Meanwhile, positive correlations between incompatible elements and SiO₂ and flat
451 correlations between other incompatible elements and Al₂O₃ would be established. However, the
452 TiO₂ content is gradually decreasing toward the chondrule margin (Fig. 2e) and negatively correlated
453 with SiO₂ content (Fig. 8d). Furthermore, the Al₂O₃ content does not exhibit a chondrule-scale
454 zonation that increases outward (flat in the upper part and symmetric zonation in the lower part, Fig.
455 2e) and shows both positive and negative correlations with SiO₂ content (Fig. 8c). Since Al and Ti
456 are both refractory elements (50% $T_c > 1582$ K) (Lodders, 2003), these two observations indicate the
457 continuous condensation of gaseous SiO onto the chondrule melt surface during olivine

458 crystallization, which diluted the concentrations of refractory elements of Al and Ti, as proposed by
459 Nagahara et al. (2008).

460 Contrary to outcomes of olivine crystallization at the surface, FeO content increases outward
461 (Fig. 2f) and shows a positive correlation with SiO₂ as well as the refractory element Al₂O₃ (Figs. 8b,
462 g), demonstrating the condensation of gaseous Fe onto chondrule melt during crystallization.
463 Diffusion of Fe from the chondrite matrix to the chondrule mesostasis during thermal metasomatism
464 is not adequate to establish this correlation between Fe and Si & Al because their diffusion rates in
465 silicate melts are different (Zhang et al., 2010). Furthermore, the MgO content, which is negatively
466 correlated with SiO₂ that follows the crystallization of olivine, displays both negative and positive
467 correlations with Al₂O₃, supporting that gaseous Mg was also condensed from ambient gas onto the
468 chondrule melt. This result is consistent with the condensation of gaseous SiO and Fe inferred
469 above because of their similar condensation temperatures (50% T_c ~1310-1336 K; Lodders, 2003).
470 The discontinuous layer of olivine at the surface of chondrules MET 00526_A2 (Fig. 1b), MET
471 00526_A1 (Fig. 1e), and NWA 8276_A2 (Fig. 2c), which is in contrast with the skeletal olivine grains
472 in the chondrules' interior, could also be resulted from condensation of gaseous Mg and SiO onto
473 chondrule melt that induced the crystallization of olivine at the surface (Nagahara et al., 2008;
474 Libourel and Portail, 2018). Minor elements V, Cr, and Mn display weak symmetric zonation across
475 the upper mesostasis of NWA 8276_A2 (Fig. S1), which can be explained by olivine crystallization at
476 the chondrule center and upper margin. Furthermore, they do not show positive correlations with the
477 refractory oxide Al₂O₃ (Fig. S2), arguing against the condensation of gaseous V, Cr, and Mn onto
478 chondrule melts, though their 50 % T_c of Cr and Mn are lower than Mg and SiO (<1300 K; Lodders,
479 2003). The nearly absent of condensation of V, Cr, and Mn gaseous species onto the chondrule
480 melts is interpreted to result from their low partial pressures in the ambient gas.

481 Glassy mesostasis in NWA 7731_A1 shows localized symmetric zonation of MgO, Al₂O₃,
482 SiO₂, and TiO₂ between dendritic high-Ca pyroxene bars (segments I, II, and IV), which could be
483 well explained by the crystallization of high-Ca pyroxene that consumed CaO, MgO, SiO₂, Al₂O₃,
484 TiO₂, and FeO, and concentrated Na₂O, SiO₂, Al₂O₃, and FeO (Fig. 3e) in the adjacent melt (bulk

485 melt is richer in SiO_2 , Al_2O_3 , and FeO than high-Ca pyroxene, ~50 wt% vs. 45 wt%, ~19 wt% vs. 15
486 wt%, and 0.11 wt% vs. 0.15 wt%, respectively) (Jones and Layne, 1997). Negative correlations
487 between MgO and TiO_2 vs. SiO_2 and positive correlations between Al_2O_3 and FeO vs. SiO_2 , as well
488 as FeO and Na_2O vs. Al_2O_3 support this conclusion (Fig. 8). The Na_2O and FeO contents display
489 chondrule-scale zonation that increases outward from the center (Fig. 3f), which haven't been
490 disturbed by the crystallization of high-Ca pyroxene. The zonation profiles are concentric, with the
491 peripheric ~100 μm layer being significantly richer in Na_2O compared to its interior and no significant
492 Na-enrichments (small spikes) near interior cracks (Fig. 3d), indicating the condensation of gaseous
493 Na and Fe onto the chondrule surface during crystallization. There is no clear evidence for gaseous
494 Mg and SiO condensation onto the chondrule melt from the elemental distributions and correlations,
495 while they must have condensed too because of their similar 50% T_c to Fe (Lodders, 2003). The lack
496 of such evidence could be due to the overprint of the crystallization effects of high-Ca pyroxene onto
497 the condensation effects of Mg and SiO, where high-Ca pyroxene crystallized as a metastable phase
498 from a highly undercooled melt (Faure and Tissandier, 2014). This overprint process occurred at a
499 lower temperature, under which the condensation of most gaseous species (like Mg, SiO, and Fe)
500 ceased, different from the case of NWA 8276_A2 that olivine crystallization and gaseous
501 condensation occurred contemporarily. The crystallization of high-Ca pyroxene disturbed the
502 distributions of MgO and SiO_2 established by condensation, but weakly affected those for Na_2O and
503 FeO , because the latter two are minor elements in high-Ca pyroxene.

504 4.2.2 Oxygen isotope evidence for gas-melt interactions

505 The plagioclase-bearing ARC MET 00526_A2 shows $\delta^{18}\text{O}$ heterogeneity with early-
506 crystallized spinel (~−10 ‰) and olivine (~−9 ‰) being more negative compared to late-crystallized
507 high-Ca pyroxene (~−8 ‰) and plagioclase (~−8 ‰ to −5 ‰) (Fig. 6), while their $\Delta^{17}\text{O}$ values are
508 within a small range of ~−5 ‰ to ~−3.5 ‰. Multiple analyses on spinel, olivine, and high-Ca
509 pyroxene grains show almost identical $\delta^{18}\text{O}$ values, suggesting that this heterogeneity is not related
510 to relict phases. The more variable and ^{16}O -depleted signatures of plagioclase could be due to
511 continuous exchange with ambient gas during its prolonged crystallization under a cooling rate of

512 1°C/hr in the range of 1000°C to 800°C (Wick and Jones, 2012) or with fluid during asteroidal
513 metasomatism as suggested by the common occurrence of scapolite (Fig. 1c). For spinel, olivine,
514 and high-Ca pyroxene that having sluggish oxygen isotope self-diffusion rates (Houlier et al., 1988;
515 Ryerson and McKeegan, 1994), they show <1 ‰ variability in $\Delta^{17}\text{O}$ (not clearly resolved) and ~2 ‰
516 variability in $\delta^{18}\text{O}$. Their $\delta^{18}\text{O}$ and $\delta^{17}\text{O}$ are distributed along a slope ~0.94 line, suggesting that this
517 $\delta^{18}\text{O}$ heterogeneity is mainly mass-independent instead of being mass-dependent relating to
518 chondrule melt evaporation (Kita et al., 2010; Piralla et al., 2021). If plagioclase is included in the
519 regression, the slope becomes ~0.87, clearly differing from the slope ~0.52 mass-dependent
520 fractionation line. Thus, we consider that the apparent correlation between $\delta^{18}\text{O}$ values and the
521 mineral crystallization sequence was most likely established by oxygen isotope exchange between
522 chondrule melt and ambient gas during its crystallization. Similarly, three analyses on the glassy
523 mesostasis of NWA 8649_A1 show $\Delta^{17}\text{O}$ from -4 ‰ to -1 ‰, with $\delta^{18}\text{O}$ and $\delta^{17}\text{O}$ distributing along a
524 slope ~1 line over a range of ~5 ‰ (Fig. 6). Assuming the glassy mesostasis in NWA 8649_A1 and
525 those in FMCs from LL3.0-3.1 chondrites were exchanged by the same fluid, the regression slope
526 derived from the former would be shallower than ~0.8 derived from the latter (Kita et al., 2010),
527 which is not the case for NWA 8649_A1. Furthermore, the center analysis is more ^{16}O -enriched than
528 the peripheral ones. These observations suggest that this heterogeneity is most likely the state of
529 the gas-melt exchange process frozen by the rapid cooling of this small droplet (~200 μm). If it is true,
530 this chondrule melt must have remained molten for a very short period because the oxygen isotope
531 homogenization timescale for a ~200 μm basaltic droplet at 1450 °C is ~0.4 hours (Lesher et al.,
532 1996). Moreover, it is also possible that the plagioclase of MET 00526_A2 and glassy mesostasis of
533 NWA 8649_A1 experienced partial oxygen isotope exchange with fluids during asteroidal
534 metasomatism and resulted in their internal $\delta^{18}\text{O}$ heterogeneity (see section 4.3.1).

535 The dependence of ^{16}O enrichment on the degree of melting (porphyritic ones are richer in
536 ^{16}O than nonporphyritic ones) found in ARCs by Russell et al. (2000) is inconsistent with the
537 observations of this study. The nonporphyritic ARCs NWA 7731_A1 ($\Delta^{17}\text{O}$: -3.6 ‰) and NWA
538 8649_A1 ($\Delta^{17}\text{O}$: -4 ‰ to -1 ‰) are more ^{16}O -enriched than the BO ARC NWA 8276_A1 ($\Delta^{17}\text{O}$: -

539 0.8 ‰) and the PO ARC NWA 8276_A2 ($\Delta^{17}\text{O}$: -0.3 ‰), but like the POP ARC MET 00526_A1
540 ($\Delta^{17}\text{O} \sim -3$ ‰). Instead, we believe that the oxygen isotope ratios of ARCs are highly dependent on
541 the degree of oxygen isotope exchange with ^{16}O -poor ambient gas in addition to their precursors
542 (initial melts).

543 **4.3 Parent body processes of OC ARCs**

544 **4.3.1 Aqueous alteration**

545 Contrary to the conventional view of dry conditions, evidence of aqueous alteration has been
546 primarily found in the most primitive OC chondrite Semarkona (LL3.00), including the presence of
547 phyllosilicates and carbonates in chondrule mesostasis and inter-chondrule matrix (Hutchison et al.,
548 1987; Alexander et al., 1989, 2015; Grossman and Brearley, 2005), “bleached chondrules”
549 (mesostasis in their outer portions have been bleached and left a porous zone depleted in alkalis
550 and Al) (Grossman et al., 2000), carbide-magnetite assemblages (Krot et al., 1997), “zoned
551 chondrules” (“sub-parallel” zonation profiles of moderately volatile elements and Ca in their
552 mesostasis) (Grossman et al., 2002), oxidized iron and iron-rich olivine in matrix and/or
553 microchondrules (Dobrică et al., 2019; Dobrică and Brearley, 2021), and disturbed Na and Ca
554 zonation in plagioclase (Lewis and Jones, 2019).

555 Aqueous alteration features have also been identified among the six ARCs of this study,
556 which are from OCs with low petrologic types (3.05) slightly more metamorphosed than Semarkona
557 (Siron et al., 2022). The occurrence of scapolite within the mesostasis, along the boundaries of
558 phenocrysts, and especially at the lower center of the plagioclase-bearing ARC MET 00526_A2 (Fig.
559 1c) suggests that it is a secondary product of plagioclase via aqueous alteration, where Na_2O , K_2O ,
560 Cl , and FeO were introduced, and CaO was leached out. An LA-ICP-MS analysis on the scapolite-
561 rich region shows a significant depletion of RLEs with strong enrichments of Sr, Ba, and Eu (Fig. 5d),
562 supporting the hypothesis that its parental phase was plagioclase. Furthermore, the hydrated portion
563 (Fig. 2) of NWA 8276_A2 is depleted in Na_2O , CaO , and Al_2O_3 and enriched in FeO , NiO , and K_2O
564 compared to interior clean glassy mesostasis, which is similar to the bleached chondrules described

565 by Grossman et al. (2000). Its interior mesostasis shows complementary and irregular profiles of
566 Na₂O and CaO (Fig. 2f) and a wide concentration range of Rb (0-4 × CI; Fig. 5b), suggesting that
567 CaO was replaced by Na₂O and the initial distributions of Na₂O and Rb were disturbed by aqueous
568 alteration. Meanwhile, the distributions of Al₂O₃, MgO, SiO₂, TiO₂, and FeO established by olivine
569 crystallization and gas-melt interactions remain largely intact because of their possible slower
570 diffusion rates in silicate glasses assuming their relative diffusivities are comparable to those in
571 silicate melts (Zhang et al., 2010). The oxygen isotope ratios obtained from its mottled mesostasis
572 are significantly enriched in ¹⁸O and ¹⁷O compared to clean regions (~4 ‰ vs. 0 ‰ and ~3 ‰ vs.
573 0 ‰, respectively; $\Delta^{17}\text{O}$: +0.6‰ vs. -0.3‰; Table 3), supporting that a fluid with heavy oxygen
574 isotopes has altered its composition.

575 4.3.2 Implication for Na-Al-rich chondrules in metamorphosed OCs

576 In metamorphosed OCs, thermal metamorphism was found to be assisted by fluid that
577 resulted in the formation of nepheline, sodalite, scapolite, amphibole, carbide-magnetite
578 assemblages, phosphate, and K-feldspar (Rubin and Grossman, 1985; Bridges et al., 1997; Krot et
579 al., 1997; Russell et al., 2000; Dobrică and Brearley, 2014; Jones et al., 2014; Lewis et al., 2022).
580 Plagioclase is weakly resistant to fluid alteration and could be replaced by sodalite/scapolite in types
581 3.2-3.6, nepheline in types 3.4-3.9, and albite in types 3.6-5 OCs (Lewis et al., 2022). Therefore,
582 previous studies on ARCs from metamorphosed type 3.1-4 OCs found plagioclases mostly altered to
583 nepheline; however, glassy mesostasis, which is more easily altered by fluids than plagioclase, was
584 found to be abundant and enriched in Na-Al (Bischoff and Keil, 1984; Krot and Rubin, 1994; Russell
585 et al., 2000; MacPherson and Huss, 2005; Jiang et al., 2015; Ebert and Bischoff, 2016; Varela and
586 Zinner, 2018; Ebert et al., 2022).

587 The Na-Al-rich chondrules are thought to have formed by remelting mixtures of a refractory
588 CAI/AOA-like component and a nepheline-rich component in closed systems with high Na partial
589 pressure (Bischoff and Keil, 1984; Krot and Rubin, 1994; Ebert and Bischoff, 2016). This hypothesis
590 is inconsistent with the open system behaviors evidenced by major element distributions and

591 correlations and oxygen isotope signatures of ARCs in Semarkona (Nagahara et al., 2008) and OCs
592 of this study (section 4.2). It also contradicts with interpretations of post-olivine crystallization entry of
593 Na into chondrule melt as suggested by the high precision EPMA analyses of sodium in olivines of
594 Na-Al-rich chondrules (Ebert et al., 2022). Instead, we argue that they are most likely
595 metamorphosed Ca-Al-rich chondrules since (i) the two have almost identical textures and mineral
596 assemblages (glassy mesostasis with dendritic high-Ca pyroxene and skeletal olivine); (ii) the
597 relative abundance of the two correlates with the petrologic type of host meteorites, with the former
598 (Na-Al-rich) being almost absent in pristine OCs (3.05, this study), coexisting with the latter in mildly
599 metamorphosed OCs (3.1-3.2; Ebert et al., 2022), and dominating in moderately metamorphosed
600 (3.3-4) OCs; (iii) the compositional differences between the two can be explained by substitutions of
601 Na and K for Ca and of Fe for Mg during thermal metamorphism (Huss et al., 2006), i.e., the glassy
602 mesostasis in the former are significantly richer in Na_2O (4-16 wt% vs. <1 wt%), K_2O (<2.5 wt% vs.
603 ~0 wt%), FeO (0.7-5.6 wt% vs. <0.5 wt%) and poorer in CaO (0-15 wt% vs. 15-20 wt%) and MgO (0-
604 11 wt% vs. 4-11 wt%), and the phenocrysts (spinel, olivine, and low-Ca pyroxene) are iron-rich (up
605 to 32 wt%) (Krot and Rubin, 1994; MacPherson and Huss, 2005; Jiang et al., 2015; Ebert and
606 Bischoff, 2016; Ebert et al., 2022), compared to the latter; and (iv) the majority of mesostasis in the
607 former are significantly ^{16}O -depleted (Ebert et al., 2022), while those in the latter are mostly not, as
608 compared to their coexisting phenocrysts, suggesting that the oxygen isotope signatures in the
609 former were modified by asteroidal fluids.

610 While abundant evidence exists for transforming Ca-Al-rich chondrules to Na-Al-rich
611 chondrules by asteroidal metasomatism, here are three major difficulties in understanding how these
612 textures have been preserved. First, how to prevent glassy mesostasis from bleaching during
613 aqueous alteration? This can be explained by the low water-rock ratio of the OC parent bodies (e.g.,
614 Alexander et al., 1989), where the metasomatism effects were subtle and localized. Previous studies
615 on Semarkona chondrules found mainly the outer zones of radial pyroxene and cryptocrystalline
616 chondrules were severely bleached (Grossman et al., 2000), while the porphyritic chondrules were
617 bleached randomly to varying degrees (Lewis and Jones, 2019). Furthermore, metasomatism

618 products of Na-Cl-rich phase (Na-Cl-rich glass, nepheline, sodalite, and scapolite) were identified in
619 only ~7-8 % chondrules in metamorphosed OCs Chainpur (LL3.4) and Parnallee (LL3.6) (Bridges et
620 al., 1997).

621 Second, how to preserve the glassy mesostasis of OC ARCs from devitrification during
622 progressive thermal metamorphism? It is known that isotropic glass is less common in chondrules
623 from OC>3.4 meteorites (metamorphic temperature >400-600°C) (Huss et al., 2006), but the glassy
624 mesostasis in ARCs from type 3-4 OCs remains mostly vitreous with minor peripheral zones being
625 devitrified (Krot and Rubin, 1994; MacPherson and Huss, 2005; Jiang et al., 2015; Ebert and
626 Bischoff, 2016). We attribute the survival of glassy mesostasis in ARCs to their Ca-Al-rich
627 compositions, where the Tg of a calcium aluminosilicate glass with similar composition (Al/Ca ratio
628 ~1, SiO₂ ~50 wt%) is ~800°C (Shelby, 1985). This Tg, while it is pressure-dependent, is significantly
629 higher than the peak metamorphic temperature (400-600 °C) of type 3-4 OCs but lower than that of
630 type 4-6 OCs (<950 °C), matching well with the absence of glassy ARCs in OCs with petrologic
631 type >4 (Krot and Rubin, 1994; MacPherson and Huss, 2005; Jiang et al., 2015; Ebert and Bischoff,
632 2016).

633 Third, what are the sources of sodium in Na-Al-rich chondrules? It is known that sodium
634 could be leached from the mesostasis and plagioclase of ARCs and FMCs by low-pH fluids at the
635 earliest stage of thermal metamorphism (petrologic type ≤3.1) (Grossman et al., 2000; Lewis et al.,
636 2022), resulting in the elevation of pH in the fluids that contributed to the formation of feldspathoids
637 (mainly nepheline) during prograde thermal metamorphism (petrologic type 3.1-3.9) (Lewis et al.,
638 2022). At this stage, the highly mobile Na in the fluids could have been reincorporated by the ARCs,
639 either by pooling in the bleached glass or substituting Ca in unbleached ones, followed by
640 dehydration due to the loss of the fluid on the asteroid through peak metamorphism. The high
641 normative albite + nepheline compositions for the mesostasis of Na-Al-rich chondrules (Ebert and
642 Bischoff, 2016), as well as the occurrence of Ca-rich rim around a Na-Al-rich chondrule (Ebert et al.,
643 2022), are consistent with the hypothesis that they were formed by bleaching Ca-Al-rich chondrules
644 followed by feldspathoid crystallization and dehydration during prograde thermal metamorphism.

645 **4.4 Precursors of OC ARCs**

646 **4.4.1 Implications from bulk major element abundances**

647 Bulk compositions of OC ARCs are significantly enriched in Al_2O_3 , TiO_2 , and CaO compared
648 to FMCs (Fig. 9a), explicitly suggesting a refractory component in their precursors. On the ternary
649 diagram of Ca_2SiO_4 – Mg_2SiO_4 – Al_2O_3 projected from spinel (Fig. 10), the OC ARCs of this study are
650 richer in MgO and poorer in CaO compared to the pyroxene-anorthite-rich CAIs (igneous type C
651 CAIs in CVs and condensate CAIs in COs and Acfer 094) and fine-grained spinel-rich CAIs. This
652 implies that the two types of CAIs could be among their precursors, but simply remelting them
653 couldn't produce OC ARCs; therefore, a nonrefractory source is needed. Because OC ARCs are Fe-
654 poor with minor or free of Fe-Ni metals and iron sulfides, the nonrefractory component could either
655 be (i) the precursor of type I chondrules ("CAI-FMC hybrids," MacPherson and Huss, 2005); or (ii)
656 the ambient gas, i.e., gaseous Mg and SiO condensed onto the CAI melts and modified their bulk
657 compositions ("CAI-Gas model"; Nagahara et al., 2008). Along with four Semarkona and three NWA
658 7402 (LL3.1) ARCs from the literature (Rubin, 2004; MacPherson and Huss, 2005; Nagahara et al.,
659 2008; Ross et al., 2017), the ARCs from OCs with petrologic type <3.1 are distributed along the
660 anorthite-forsterite join toward the forsterite apex of the ternary diagram (Fig. 10), rather than
661 extending from the two types of CAIs to type I chondrules, supporting that "CAI-Gas" model. On the
662 diagram of SiO_2 – CaO – Al_2O_3 – MgO (Fig. 10 of Nagahara et al., 2008), these OC ARCs follow a
663 nearly linear trend that intersects the MgO - SiO_2 axis at an atomic ratio of 3:2 to 2:1 (forsterite, Fig.
664 S4), indicating that the gaseous Mg and SiO added to the chondrule melt was at an atomic ratio of
665 ~3:2 to ~2:1. Furthermore, the bulk compositions of metamorphosed OC ARCs, corrected for the Fe-
666 Mg and (Na, K)-Ca exchanges due to thermal metasomatism, overlap largely with pyroxene-
667 anorthite-rich CAIs and partially with fine-grained spinel-rich CAIs (Fig. 10). Like those from OCs
668 with petrologic type <3.1, their bulk compositions are well-distributed along the An-Fo join, as
669 illustrated by their 2D Kernel density distributions of the three apexes (Forsterite, Corundum, and
670 Larnite) calculated from their corrected bulk compositions (Fig. S3).

671 4.4.2 Implications from bulk trace element abundances

672 Bulk RLE (Zr, Hf, Sc, Y, Ti, Sr, Ba, and REEs, $\sim 7-15 \times \text{CI}$) abundances of OC ARCs in this
673 study are significantly enriched compared to FMCs ($1-3 \times \text{CI}$ for Sc, Hf, La, Sm, Eu, Tb, Yb, and Lu,
674 determined by INNA) (Gooding et al., 1980; Grossman and Wasson, 1982, 1983; Kurat et al., 1984;
675 Rubin and Pernicka, 1989; Swindle et al., 1991), suggesting a refractory RLE-rich component in their
676 precursors, most likely CAIs (tens of CI, e.g., Grossman, 1973; Mason and Martin, 1977; Mason and
677 Taylor, 1982). The findings of group II (occasionally with negative Sm anomaly) and III REE patterns
678 in five out of nine OC ARCs (Fig. 5e) studied by Ebert and Bischoff (2016), which were established
679 during condensation from a gas of near solar composition in the earliest epoch of the protoplanetary
680 disk (Boynton, 1975; Davis and Grossman, 1979), undoubtedly indicate that the refractory
681 component is CAIs. The OC ARCs of this study all display unfractionated (group I, Fig. 5a-d) REE
682 patterns similar to those reported in MacPherson and Huss (2005), suggesting that RLEs in the
683 majority of OC ARCs (17 out of 22) are unfractionated, in stark contrast with ARCs in CV chondrites
684 (7 out of 11 have fractionated group II-like or III REE patterns; Wakaki et al., 2013; Zhang et al.,
685 2014; Zhang et al., 2020b). This observation rules out the possibility of fine-grained spinel-rich CAIs,
686 which typically have fractionated group II REE patterns (e.g., Krot et al., 2004b), as the refractory
687 component in the precursors of OC ARCs.

688 Bulk Cr and Mn abundances of OC ARCs in this study are depleted to varying degrees (Cr =
689 $0.2-0.6 \times \text{CI}$, Mn = $0.1-0.4 \times \text{CI}$, Figs. 5) compared to OC FMCs ($\sim 1-3 \times \text{CI}$) (Gooding et al., 1980;
690 Grossman and Wasson, 1982, 1983; Kurat et al., 1984; Rubin and Pernicka, 1989; Swindle et al.,
691 1991). In section 4.2.1, elemental distribution and intercorrelations demonstrated that almost no
692 recondensation of Cr and Mn occurred during melt crystallization. Thus, the depletion of Cr and Mn
693 in OC ARCs indicates a Cr-Mn-depleted component in their precursors, most likely CAIs (Cr $< 0.7 \times$
694 CI and Mn $< 0.5 \times \text{CI}$; Grossman, 1973; Grossman and Ganapathy, 1975; Grossman and Ganapathy,
695 1976b, a; Sylvester et al., 1993).

696 Since REE-unfractionated CAIs and OC FMCs both exhibit limited ranges of REE
697 abundances ($\sim 18 \pm 4 \times \text{CI}$ in CAIs and $1.8 \pm 1 \times \text{CI}$ in FMCs) (Martin and Mason, 1974; Wänke et al.,

698 1974; Mason and Martin, 1977; Gooding et al., 1980; Grossman and Wasson, 1982, 1983; Mason
699 and Taylor, 1982; Kurat et al., 1984; Rubin and Pernicka, 1989; Swindle et al., 1991; Sylvester et al.,
700 1993; Huang et al., 2012; Davis et al., 2018; Torrano et al., 2019, 2023), it is possible to estimate the
701 portion of CAI component in the precursors of OC ARCs based on their REE abundances. Rough
702 calculations suggest that the CAI component accounts for ~30-100 % of their precursors (Table 2,
703 Fig. 9b); the wide range is due to the relatively large uncertainty of REE abundance of the CAI
704 endmember. Applying the same calculation to REE-unfractionated OC ARCs in the literature
705 (Chainpur 5674-2-1 & 1251-14-2 & 1251-3-1, Inman 5652-1-1, and Hammadah al Hamdra 335-3)
706 (MacPherson and Huss, 2005; Ebert and Bischoff, 2016) indicate a portion of CAI of ~30-100 %.
707 Furthermore, the REE abundances of Semarkona 4128-3-2, Chainpur 1251-14-1, Quinyambie 6076-
708 5-2, and Hammadah al Hamdra 335-1 (MacPherson and Huss, 2005; Ebert and Bischoff, 2016) are
709 too low to suggest a CAI component in their precursors.

710 Utilizing the CAI portion estimated from REEs and the average compositions of type IA and
711 IAB/IB chondrules ("CAI-FMC hybrid" model; Jones and Scott, 1989; Jones, 1994) or the inferred
712 ambient gas composition (MgO:SiO~3:2 to 2:1; Nagahara et al., 2008), the major element
713 compositions of the CAIs incorporated into the precursors of OC ARCs are roughly calculated to
714 contain 15-25 wt% CaO, 15-35 wt% Al₂O₃, 0-20 wt% MgO, and 30-50 wt% SiO₂ (assume similar
715 gaseous recondensation effects for OC ARCs and type I chondrules; "CAI-FMC hybrid" model) or
716 15-25 wt% CaO, 20-35 wt% Al₂O₃, 0-15 wt% MgO, and 30-50 wt% SiO₂ (the "CAI-Gas" model). The
717 inferred composition ranges are very similar, and both significantly overlap with pyroxene-anorthite-
718 rich CAIs on the ternary diagram of Ca₂SiO₄-Mg₂SiO₄-Al₂O₃ (green and magenta zones in the inset
719 of Fig. 10), making this type of CAI to be the best candidate recycled by OC ARCs. Furthermore, to
720 avoid negative MgO and SiO₂ contents for the CAI component, the lower limit portion of the CAI
721 component is typically higher than that estimated from REE abundances (Table 2). It is noteworthy
722 that the bulk V content of OC ARCs of this study (0.3-1.9 × CI) and the literature (0.7-3.5 × CI; Ebert
723 and Bischoff, 2016) are significantly lower than other REEs (i.e., Zr, Hf, Sc, Y, Ti, Sr, Ba, and REEs,

724 ~7-15 × CI), implying a significant loss of V via unknown processes associated with chondrule
725 formation or asteroidal metamorphism.

726 4.4.3 Implications from oxygen isotopes

727 Previous studies found oxygen isotopes of OC ARCs extend from the typical oxygen isotope
728 region of FMCs toward a more ^{16}O -enriched endmember ($\delta^{18}\text{O}$ down to $-15\text{\textperthousand}$) along a line of slope
729 0.83 ± 0.09 (Russell et al., 2000) or 0.7 ± 0.1 (Jiang et al., 2015). The mixing lines are shallower than
730 the carbonaceous chondrite anhydrous minerals line (CCAM, slope = 0.941, intercept = -4.00 ;
731 Clayton et al., 1973) and the primitive chondrule minerals line (PCM, slope = 0.987, intercept = $-$
732 2.70 ; Ushikubo et al., 2012) and do not intersect with the oxygen isotope region of CAIs, indicating
733 that the OC ARCs are not simple mixtures of CAIs and FMC materials. In contrast, a recent study by
734 Ebert et al. (2022) found that the oxygen isotopes of spinel, olivine, and low-Ca pyroxene in
735 metamorphosed OC ARCs roughly follow the Young & Russell line (slope = 1.00, intercept = -1.044 ;
736 Young and Russell, 1998) or the PCM line (Ushikubo et al., 2012), and argued that their precursors
737 could be a mixture of CAIs(-like) and FMC materials. In this study, we determined oxygen isotope
738 ratios of six ARCs from type 3.05 OCs with significantly improved analytical precision ($\sim 0.2\text{-}0.5\text{\textperthousand}$ for
739 $\delta^{18}\text{O}$) and found that they distribute along a mixing line of $\delta^{17}\text{O} = (0.99 \pm 0.05) \times \delta^{18}\text{O} - (0.40 \pm 0.32)$
740 (MSWD = 2.4, based on data in Table 3 excluding altered glass, Fig. 11, named “OC ARC line”).
741 Semarkona ARC CH5 (Kita et al. 2010) is consistent with the OC ARC line and located near the
742 intersection with the terrestrial fractionation line (TFL, Fig. 11a). Unlike CCAM and PCM lines, the
743 mixing line is less well constrained (slope ranges from 0.92 to 1.04, orange shadowed region in Fig.
744 11), due to their small variations in oxygen isotopes ($\sim 10\text{\textperthousand}$ in $\delta^{18}\text{O}$).

745 The OC ARC line plots on the left (lower $\delta^{18}\text{O}$) of most OC type IAB, IB, and II chondrules
746 (Fig. 11a; except for IAB chondrules Bishnupur B12, Semarkona CH33, and NWA 7731 C5 that are
747 on the OC ARC line; Kita et al. 2010; Siron et al., 2022), suggesting that the ^{16}O -poor endmember is
748 not the precursor of these types of FMCs. In contrast, the OC ARC line intersects with type IA
749 chondrules with $\Delta^{17}\text{O} > 0\text{\textperthousand}$ (“supra-TFL” group defined by Marrocchi et al., 2024), which displays

750 large mass-dependent isotope fractionations with $\delta^{18}\text{O}$ ranging from $\sim-3\text{\textperthousand}$ to $\sim+9\text{\textperthousand}$ (Fig. 11b). For
751 type IA chondrules with $\Delta^{17}\text{O} < 0\text{\textperthousand}$ (“sub-TFL” group defined by Marrocchi et al., 2024), most are
752 consistent with the PCM line that could have recorded the oxygen isotope evolving from ^{16}O -rich to
753 ^{16}O -poor in the OC chondrule-forming regions (Kita et al., 2010; Marrocchi et al., 2024). The relict
754 olivines ($\Delta^{17}\text{O} \sim-5\text{\textperthousand}$ to $-2\text{\textperthousand}$) identified in IA chondrules by Kita et al., (2010) and Piralla et al.
755 (2021) are likely to be unmelted fragments of the pre-existing sub-TFL chondrules (Marrocchi et al.,
756 2024). As exceptions, some sub-TFL IA chondrules plot on the OC ARC line and have either lower
757 or higher $\delta^{18,17}\text{O}$ than OC ARCs of this study (Fig. 11b). Thus, as indicated by Nagahara et al.
758 (2008), OC ARCs and some type IA chondrules could have formed from common refractory and
759 ferromagnesian precursor solids and experienced open-system formation that involved evaporation
760 and re-condensation of Mg and SiO.

761 Toward the lower end, the OC ARC line intersects with the typical oxygen isotope region of
762 CAIs and AOAs on the CCAM and PCM lines (Fig. 11c), supporting the interpretation that CAIs were
763 among the precursors of OC ARCs. The two AOA-like relict olivines ($\Delta^{17}\text{O} < -15\text{\textperthousand}$) identified among
764 eleven IA chondrules by Piralla et al. (2021) are consistent with the PCM line and, within uncertainty,
765 also with the OC ARC line. The rarity of AOA-like olivines in OC FMCs suggests that AOAs and
766 possibly CAIs existed but as a minor component in the OC chondrule-forming region. Considering
767 the large portion of CAIs in their precursors (30-90%), the OC ARCs seem to be less ^{16}O -enriched,
768 as noticed by Russell et al. (2000), which could be due to the extensive gas-melt interactions that
769 significantly diluted the initial oxygen isotope ratios of the chondrule melt.

770 4.4.4 The Ti isotope conundrum

771 The involvement of CAIs in the formation of OC ARCs seems to conflict with the Ti isotope
772 signatures of Na-Al-rich chondrules from metamorphosed OCs, which are indistinguishable from OC
773 FMCs ($\varepsilon^{50}\text{Ti} \sim 0$) and much lower than that of OC CAIs ($\varepsilon^{50}\text{Ti} \sim 2-11$) and CO ARCs ($\varepsilon^{50}\text{Ti} \sim 2-15$)
774 (Ebert et al., 2018). Since Ti is an immobile element and no Ti gain/loss was observed in ARCs of
775 this study (section 4.2.1), the Ti isotope signatures of OC ARCs suggest the existence of a refractory

776 component without ^{50}Ti excess in the inner solar system as proposed by Ebert et al. (2018).
777 Combined with our discussions above, the most possible refractory components for OC ARCs are
778 pyroxene-anorthite-rich CAIs, which likely formed by gas condensation at lower temperatures near
779 the protosun (~1300-1450K; Ebel and Grossman, 2000). The component could be indigenous in the
780 inner disk, which might have been trapped in a magnetically dead zone at the midplane of the
781 protoplanetary disk before the onset of chondrule formation (Jacquet et al., 2011), or exotic from the
782 outer disk by inward drifting through the Jupiter gap (Desch et al., 2018; Haugbølle et al., 2019). In
783 this study, MET 00526_A2 appears to show mass-independent oxygen isotope fractionation and has
784 an Al-Mg age of $2.15 \pm 0.22/0.18$ Ma that is within the range of OC FMCs (~1.2-3.0 Ma; Fig. 12;
785 Hutcheon and Hutchison, 1989; Kita et al., 2000; Rudraswami and Goswami, 2007; Rudraswami et
786 al., 2008; Villeneuve et al., 2009; Mishra et al., 2010; Bolland et al., 2019; Pape et al., 2019; Siron et
787 al., 2021, 2022). If only the most recent Al-Mg data of OC FMCs were considered (~1.8-2.2 Ma;
788 Siron et al., 2021, 2022), various types of chondrules display a limited range of ~0.4 Ma and are
789 mostly indistinguishable to each other. While the inferred Al-Mg age of MET 00526_A2 overlaps with
790 the majority of FMCs studied by Siron et al., (2021, 2022) within analytical errors, data plot close to
791 the younger limit and postdate formation of a half of the anorthite-bearing chondrules occurring at
792 ~1.9 Ma (Fig. 12). Nevertheless, the internal oxygen isotope variation of MET 00526_A2 indicates
793 that it might not have been remelted/recycled after solidification (otherwise, homogenized). Thus, its
794 Al-Mg age indicates that the refractory component in its precursor must arrive at the OC regions
795 after, rather than before, the onset of the chondrule heating events.

796 It is worth noting that spinel of MET 00526_A2 does not show resolvable ^{26}Mg excess,
797 differing from the spinels in two OC FMCs (Semarkona ch10 and Piancaldoli ch8) that show large
798 excess that indicates an age of ~0.8 Ma (Piralla et al., 2023). Piralla et al., (2023) argued that Mg
799 diffusion in spinel is slower than in plagioclase, though Mg diffusion in plagioclase shows a strong
800 dependence to anorthite composition (Van Orman et al., 2014) and those for $\text{An} \geq 93$ are slower
801 than that of spinel (Sheng et al., 1992; LaTorrette and Wasserburg, 1998; Van Orman et al., 2014).
802 Plagioclase in MET 00526_A2 are anorthitic (An_{91-99}) so that it is unlikely that spinel recorded earlier

803 formation age than the isochron determined by plagioclase data. More studies are required to
804 examine spinel Al-Mg chronology from chondrules in which plagioclase preserved correlated ^{26}Mg
805 excess.

806 To further reveal the characteristics of this refractory component, more high-precision Ti
807 isotope analyses on other non-carbonaceous materials, for example, the ARCs in enstatite
808 chondrites, are necessary to confirm the universal existence of this unique refractory material in the
809 inner protoplanetary disk. Meanwhile, to eliminate the concern of the effects of thermal
810 metamorphism on the preservation of primary Ti isotope signatures, self-diffusion experiments of Ti
811 isotopes in Ca-Al-silicate glass or direct measurements of ARCs in OCs with low petrologic types are
812 needed.

813 **4.5 Formation mechanism for OC ARCs**

814 To summarize the discussions above, we outline the formation mechanism for OC ARCs as
815 follows (Fig. 13): (i) Stage I: condensation of pyroxene-anorthite-rich CAIs from nebular gas between
816 1300-1450 K near the protosun. The CAIs are characterized by solar-like ^{16}O -rich signatures,
817 unfractionated and, in rare cases, fractionated REE abundances, and absence of ^{50}Ti excess. They
818 could have been produced indigenous and then preserved in a magnetically dead zone of the inner
819 disk or transported in from the outer disk by gas drag; (ii) Stage II: Flash melting of the CAIs
820 subsequently interacted extensively with the ambient gas in the chondrule-forming region. During
821 mineral crystallization, gaseous Mg, SiO, Fe, and alkalis condensed onto the melts, and meanwhile,
822 diluted the ^{16}O -rich signatures of the initial melt by the ^{16}O -poor gas and established the correlation
823 between ^{16}O -enrichments and mineral crystallization sequence for MET 00526_A2. The atomic ratio
824 of condensed MgO and SiO was inferred to be ~3:2 (Nagahara et al., 2008) to ~2:1; and (iii) Stage III:
825 Asteroidal modifications of the mineralogy and oxygen isotopes of OC ARCs by fluids, including
826 bleaching glassy mesostasis by low-T fluids, followed by replacement of glassy mesostasis and
827 plagioclase with feldspathoids and dehydration during prograde metamorphism, transforming Ca-Al-
828 rich chondrules into Na-Al-rich chondrules.

829 Bischoff et al., (1989) first proposed that OC ARCs could be splashed melts of partially
830 molten FMCs because of their fractionated REE patterns. The splash model was argued against by
831 Ebert and Bischoff (2016) since there are no systematic size differences between the two. Marrocchi
832 et al., (2024) broaden the splash model from OC ARCs to sub-TFL IA chondrules, which appears to
833 be smaller than supra-TFL IA and other types of chondrules. The six ARCs of this study show
834 variable sizes (~1100 μm to ~200 μm) and modal mineral abundances (0-44 vol% olivine) that
835 cannot be explained by the splash model. They show flat REE patterns (Fig. 5) without depletion in
836 Mg (Fig. 9a, 0.8-2 $\times\text{CI}$), suggesting that they are not residual melts after the crystallization of olivine
837 and/or pyroxene. Furthermore, their oxygen isotopes are distributed on a line distinct from the
838 majority of OC FMCs (Fig. 11a, b), indicating that they formed from different precursors and
839 exchanged with isotopically different ambient gases. Thus, we consider the splash model to be
840 invalid for the formation of OC ARCs and possibly the sub-TFL IA chondrules studied by Marrocchi
841 et al., (2024).

842 **5. Conclusions**

843 We characterized petrography, mineralogy, bulk major and trace element chemistry, and
844 oxygen isotopes of six ARCs from OC3.05 meteorites. The plagioclase-bearing ARC was also
845 investigated with Al-Mg chronology. The findings provide better understanding of the mechanisms of
846 OC ARC formation, including their precursor compositions, gas-melt interactions, and asteroidal
847 metasomatism. Key observations and their interpretations are listed below.

- 848 (1) Chondrule-scale chemical zonation of major elements (Mg, Si, Al, Ti, Fe, and Na) and their inter-
849 element correlations in the glassy mesostasis of two ARCs demonstrated the condensation of
850 gaseous Mg, SiO, Fe, and alkalis onto chondrule melts.
- 851 (2) Bulk major element compositions of ARCs from OC<3.1 meteorites are closest to the pyroxene-
852 anorthite-rich CAIs and follow a nearly linear trend toward the Mg: SiO ~ 3:2 to 2:1 (in atomic)
853 apex, suggesting that OC ARCs could be made by varying degrees of gaseous Mg and SiO
854 condensation onto bulk melts of pyroxene-anorthite-rich CAIs. Their bulk trace element

855 concentrations display unfractionated REE patterns ($\sim 7-15 \times \text{CI}$) with varying depletions of V, Cr
856 and Mn, indicating that 30-100 % of the bulk is contributed by CAIs.

857 (3) Oxygen isotopes of the plagioclase-bearing ARC, MET 0526_A2, show ^{16}O -rich enrichments
858 following the mineral crystallization sequence, demonstrating gas-melt oxygen isotope exchange
859 during crystallization. A similar case is NWA 8649_A1, which shows $\sim 3 \text{ ‰}$ variation of $\Delta^{17}\text{O}$ in its
860 glassy mesostasis. Together with the remaining three displaying oxygen isotope homogeneity,
861 all OC ARCs of this study plot along a line of slope $\sim 0.99 \pm 0.05$ that is almost parallel to the
862 PCM line but towards higher $\delta^{17}\text{O}$. The OC ARC mixing line intersects with the CAIs/AOAs,
863 consistent with a small number of IA chondrules, but deviates from the majority of OC FMCs
864 (IAB/IB/II), suggesting a possible genetic relationship between OC ARCs and some OC type IA
865 chondrules; both could be made by oxygen isotope exchange between CAI melt and ambient
866 ^{16}O -poor gas.

867 (4) The OC ARCs of this study experienced mild low-T metasomatism, including bleaching of
868 chondrule mesostasis, replacing plagioclase with scapolite, and exchanging the mesostasis to
869 be more ^{16}O -poor. In comparison with Na-Al-rich chondrules in metamorphosed OCs, we argue
870 that these OC ARCs formed by metamorphism of Ca-Al-rich chondrules via bleaching,
871 albitization, and dehydration during prograde thermal metamorphism.

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881 **Appendix A. Supplementary material**

882 The supplementary materials are: (1) BSE images of SIMS oxygen isotope analysis pit; BSE
883 images and EDS results of SIMS Al-Mg isotope analysis pits; and optical images of LA-ICP-MS trace
884 element analysis pits; (2) EPMA analysis condition, EPMA major element data, LA-ICP-MS trace
885 element data, SIMS oxygen isotopes and Al-Mg isotopes data and related data processing; and (3)
886 Four supplementary figures relating to the element distribution & correlation and bulk compositions
887 of OC ARCs.

888 **Data availability**

889 Data are available through Mendeley Data at <https://doi.org/10.17632/p3bt2wbcmj.1>

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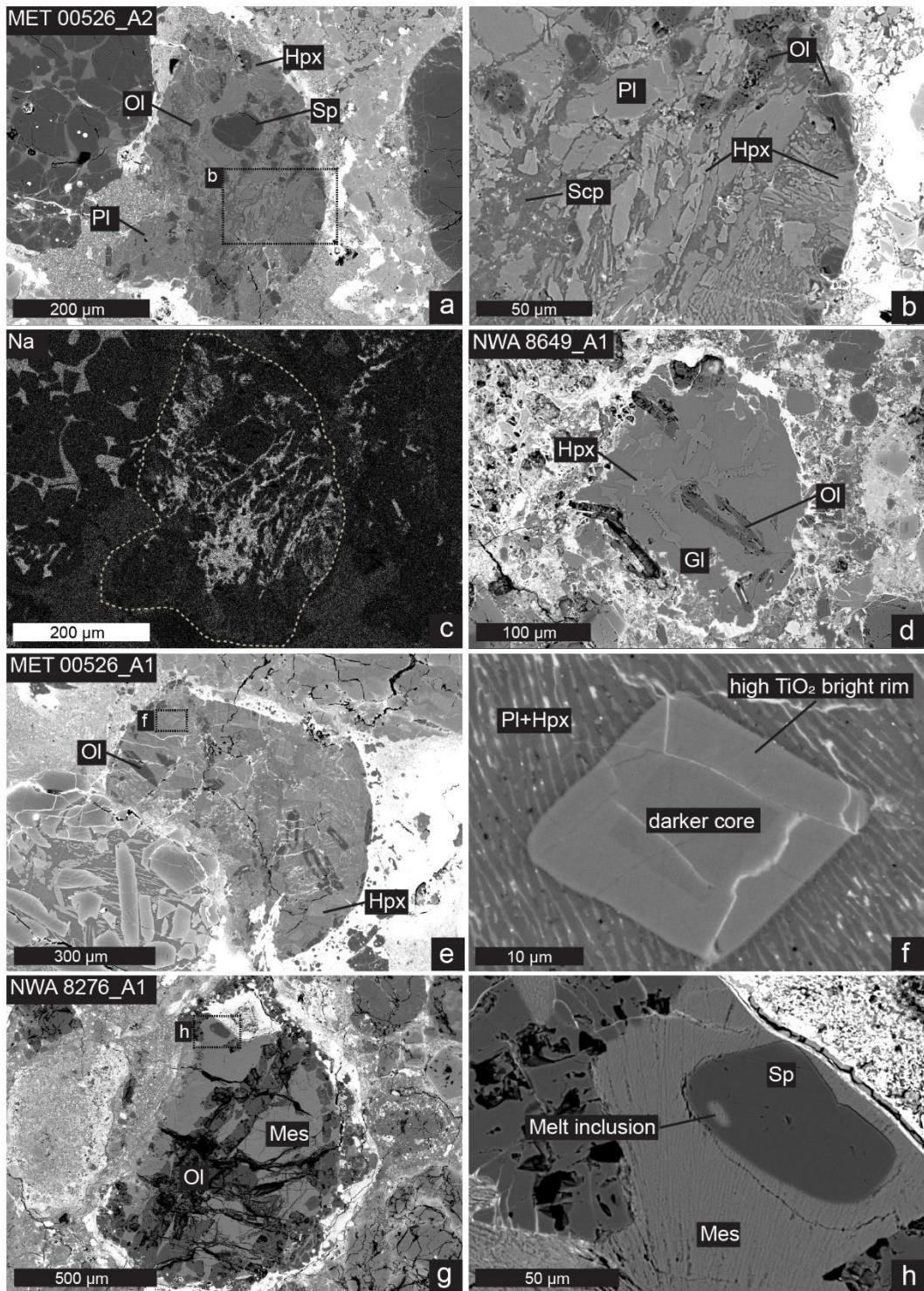
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1243 Fig. 1. Petrography of four OC ARCs in this study. (a, b) BSE images of MET 00526_A2 show it is a
 1244 plagioclase-bearing chondrule composed of spinel, olivine, plagioclase, high-Ca pyroxene, and
 1245 mesostasis of microcrystalline plagioclase and high-Ca pyroxene. Olivine occurs mainly as tablet

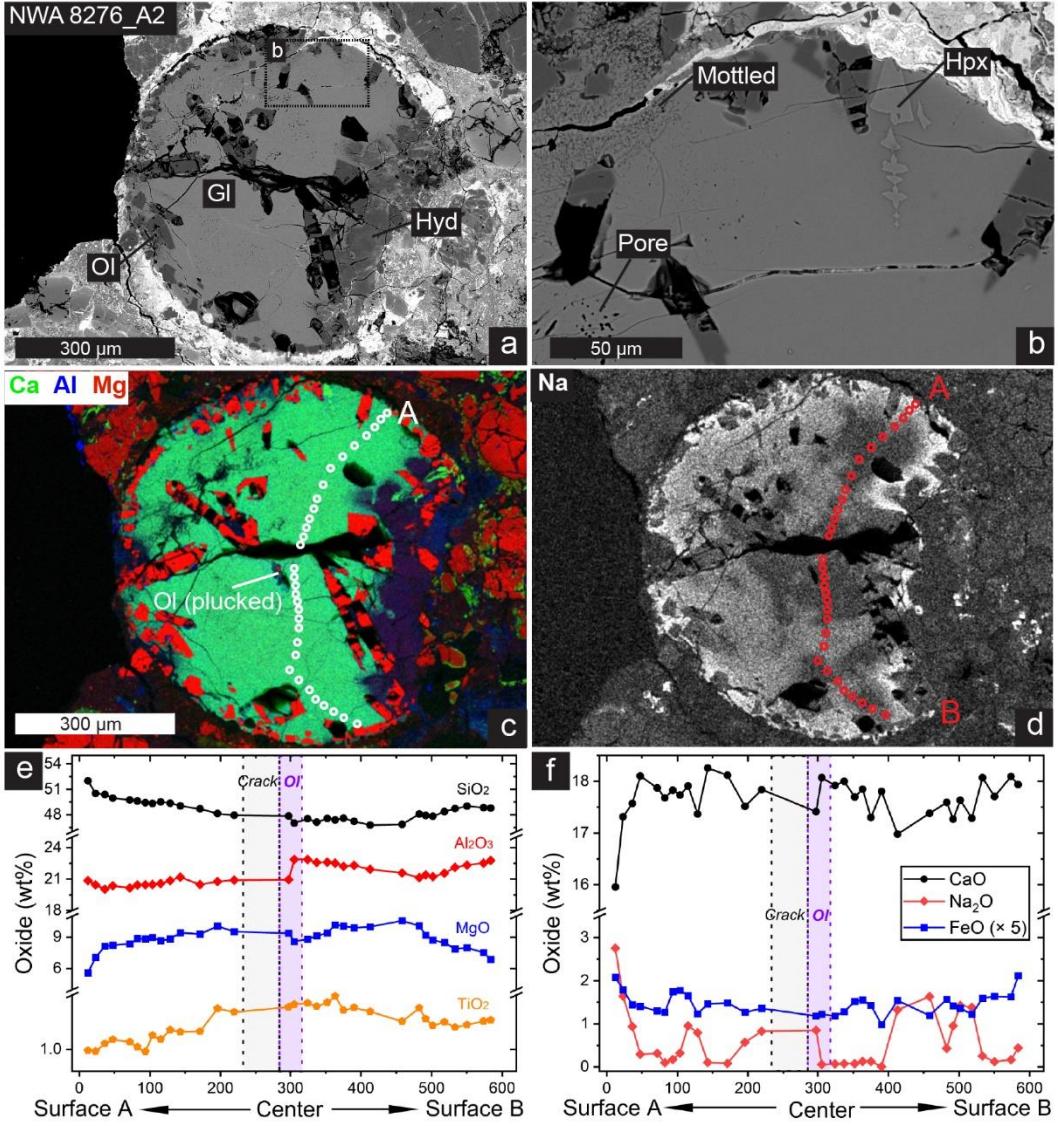
1246 crystals and, occasionally, as anhedral crystals decorating the chondrule's surface. (c) X-ray
1247 elemental map of Na of MET 00526_A2 illustrates the distribution of scapolite within this chondrule.
1248 (d) NWA 8649_A1, an ovoid glassy ARC with an apparent diameter of ~200 μm . It has skeletal
1249 olivine and dendritic high-Ca pyroxene embedded in a clean Ca-Al-rich glass. (e, f) MET 00526_A1
1250 (570 \times 300 μm), a glassy ARC with curved sides, while its Y axis was squeezed by nearby large
1251 chondrules during accretion. It contains skeletal olivine and diamond-shaped or blocky high-Ca
1252 pyroxene, with the core being darker (lower TiO_2) than the rim. Its mesostasis is an intergrowth of
1253 microcrystalline plagioclase and high-Ca pyroxene immersed with many iron oxide veins formed by
1254 terrestrial weathering. (g, h) NWA 8276_A1, the largest ARC (1100 \times 700 μm) of this study, is oval-
1255 shaped and composed mainly of olivine and mesostasis, with minor amounts of spinel and high-Ca
1256 pyroxene. It has a barred-olivine texture with parallel olivine bars located at the center of this
1257 chondrule. Blocky olivine crystals were observed to be aligned along the chondrule surface. Its
1258 mesostasis was slightly devitrified as feathery crystals likely plagioclase and high-Ca pyroxene.
1259 Abbreviations: Sp = spinel; OI = olivine; Hpx= high-Ca pyroxene; PI = plagioclase; Scp = Scapolite;
1260 GI = glass; Mes = mesostasis.

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1266 Fig. 2. Petrography and elemental distribution of the glassy ARC NWA 8276_A2. (a) BSE image
 1267 shows a nearly round chondrule with an apparent diameter of ~580 μm , dominated by olivine and
 1268 glassy mesostasis. Olivine is tablet and preferentially located at the margin of the chondrule with a
 1269 long axis parallel or normal to the surface. Some interior olivine crystals were partially or completely
 1270 plucked during sample preparation and left prismatic-shaped cavities. (b) An enlarged view shows a
 1271 dendritic high-Ca pyroxene at the surface. Micron-sized pores and mottled regions ("Mottled") were
 1272 randomly distributed in the mesostasis. (c) A combined X-ray elemental map (Ca: green, Al: blue,
 1273 Mg: red) shows a hydrated phase (analysis total ~70 wt%) at the right side of this chondrule, likely
 1274 formed by leaching the glassy mesostasis. (d) An X-ray elemental map of Na shows Na-rich

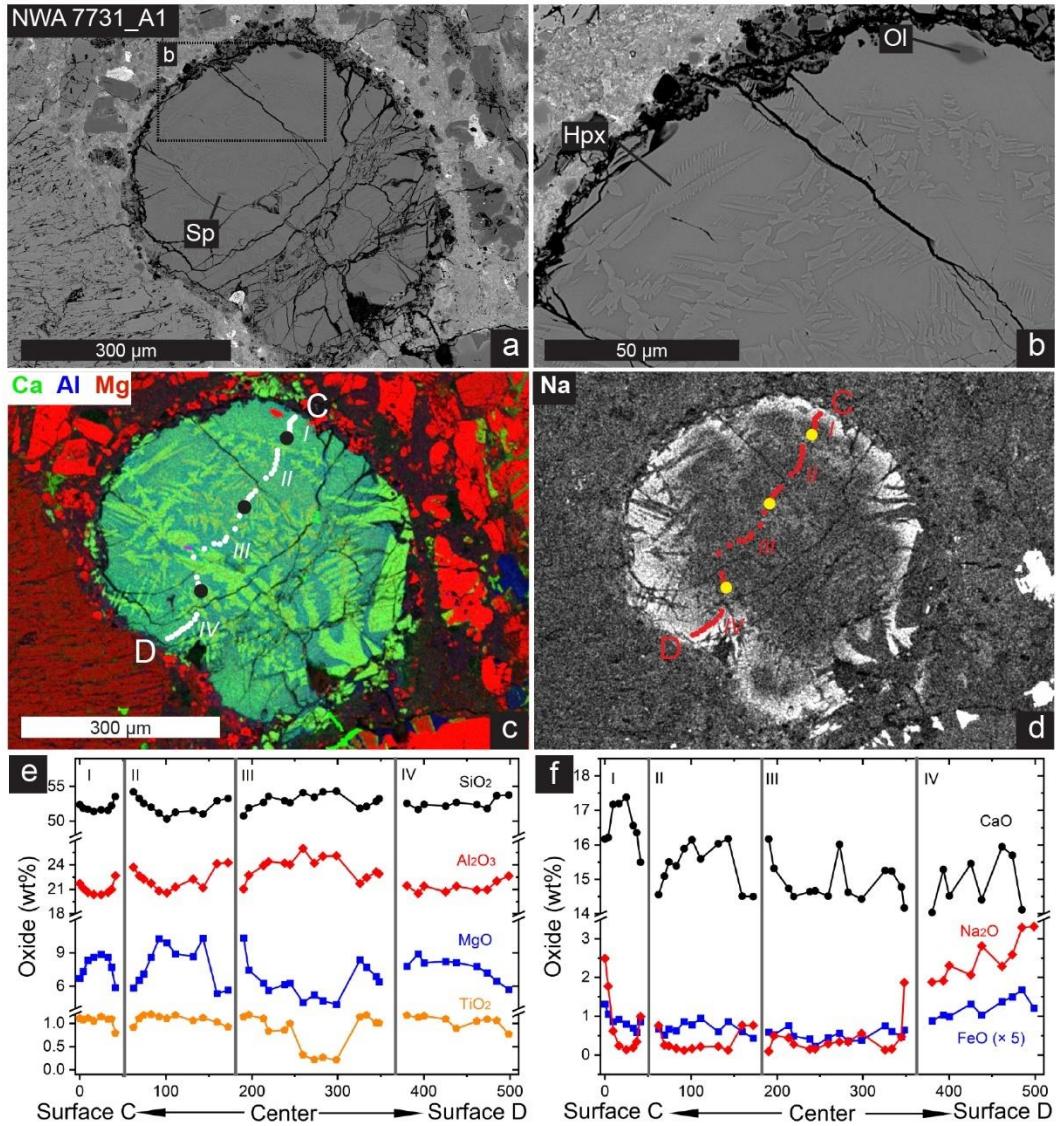
1275 domains are bracketed by Na-poor domains, and the interface between the hydrated phase and
1276 inferior glass mesostasis is significantly Na-rich. (e) Transverse EPMA analysis (A-B) shows
1277 chondrule-scale concentric zonation of MgO, SiO₂, and TiO₂ in the glassy mesostasis and symmetric
1278 zonation of Al₂O₃ in the lower half of this chondrule. It is noted that the abrupt increase of Al₂O₃ and
1279 TiO₂ and decrease of MgO and SiO₂ observed at the center resulted from a nearby olivine
1280 crystallization (indicated “plucked” in panel c). (f) EPMA line profiles of Na₂O, CaO, and FeO. The
1281 CaO profile is irregular and complementary to the Na₂O profile. The FeO profile is concentrically
1282 zoned like SiO₂. To plot the FeO profile in the same figure, it was shown as five times its actual
1283 contents (< 0.5 wt%). Abbreviations: Hyd = hydrated phase; Mottled = mottled region in glassy
1284 mesostasis; others are the same as in Fig. 1.

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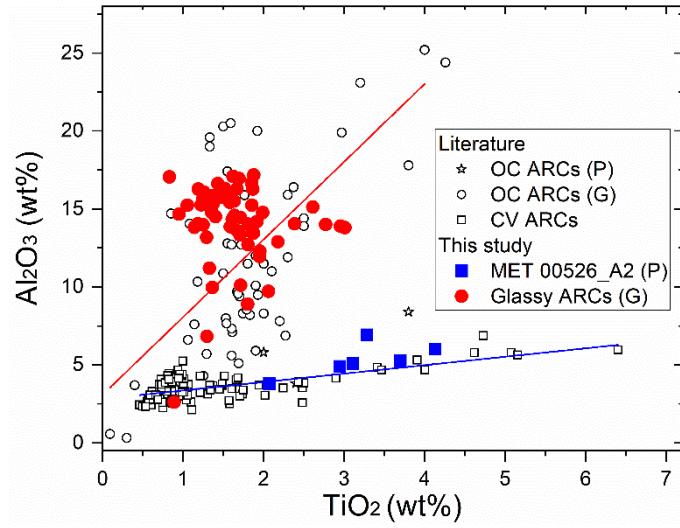


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1290 Fig. 3. Petrography and elemental distribution of the glassy ARC NWA 7731_A1. (a) BSE image
 1291 shows it is round with an apparent diameter of ~440 μm . The periphery of this chondrule is almost
 1292 vacant with minor amounts of high-Ca pyroxene and olivine fragments, which could be remnants of
 1293 dissolving glassy mesostasis by asteroidal fluids. A skeletal-shaped cavity at its lower right could be
 1294 a relic of an olivine crystal. (b) An enlarged view shows dendritic high-Ca pyroxene and clean glassy
 1295 mesostasis. A blocky olivine grain was found at its upper margin. (c) A combined X-ray elemental
 1296 map of Ca (green), Al (blue), and Mg (red) shows the dendritic shape of high-Ca pyroxene and its
 1297 distribution. A tiny spinel grain (purple) was identified at the center. (d) X-ray elemental map of Na
 1298 shows concentric zonation of Na₂O, with the outer ~100 μm region being richer in Na₂O than the

1299 interior ~300 μm . Several slightly brighter subregions were observed in the interior. (e) Profiles of
1300 SiO_2 , Al_2O_3 , MgO , and TiO_2 show localized chemical zonation in segments I, II, and IV, which were
1301 separated by high-Ca pyroxene bars, while segment II that across a coarse high-Ca pyroxene shows
1302 opposite zonation. (f) Profiles of CaO , Na_2O , and FeO . CaO shows irregular distributions, while
1303 Na_2O (with small spikes) and FeO show concentric zonation. Abbreviations are the same as in Fig. 1.

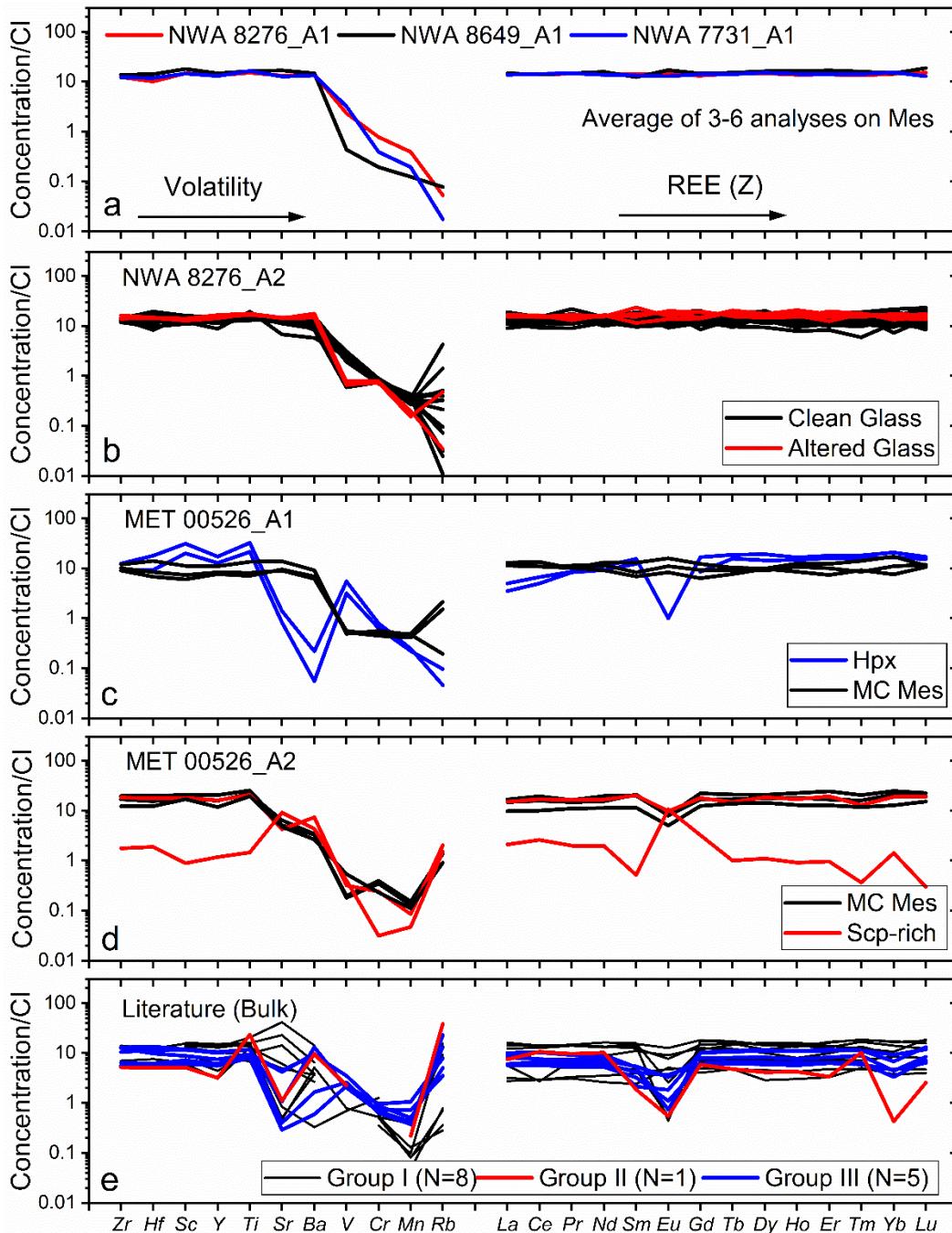
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1306 Fig. 4. Al_2O_3 vs. TiO_2 contents in high-Ca pyroxene of the six ARCs studied. High-Ca pyroxene from
 1307 the plagioclase-bearing ARC ("P") MET 00526_A2 follows a linear trend with a slope of ~ 0.5 like
 1308 other plagioclase-bearing OC ARCs (MacPherson and Huss, 2005) and porphyritic ARCs in CV
 1309 chondrites (Zhang et al., 2020b). In contrast, high-Ca pyroxene from five glassy ARCs ("G") of this
 1310 study has much higher Al_2O_3 that shows poor correlation with its TiO_2 contents, like other glassy OC
 1311 ARCs in the literature (Krot and Rubin, 1994; MacPherson and Huss, 2005; Jiang et al., 2015; Ebert
 1312 and Bischoff, 2016; Ebert et al., 2022).

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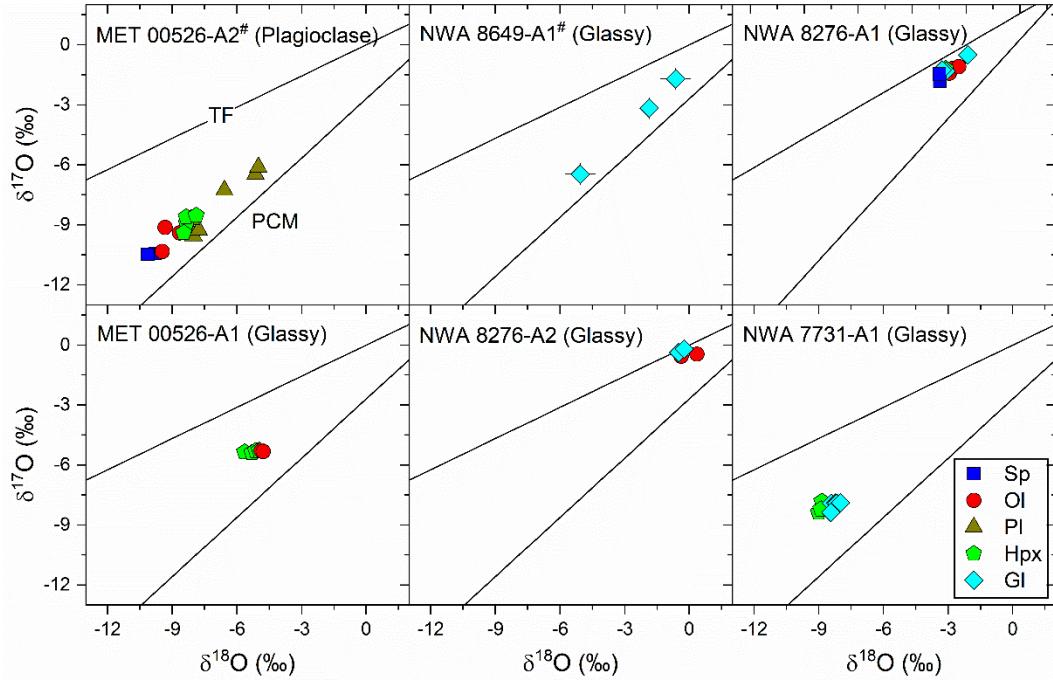
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1315 Fig. 5. Trace element abundances of mesostasis and/or high-Ca pyroxene in the six OC ARC
 1316 studied. REEs are plotted separately from other trace elements. From left to right, other elements
 1317 follow their order of volatility, and REEs follow their order of atomic number (Z). (a) Three to six
 1318 analyses of NWA 8276_A1, NWA 8649_A1, and NWA 7731_A1 show uniform trace element
 1319 concentrations and, for clarity, only average values are plotted. They show unfractionated RLE (Zr,

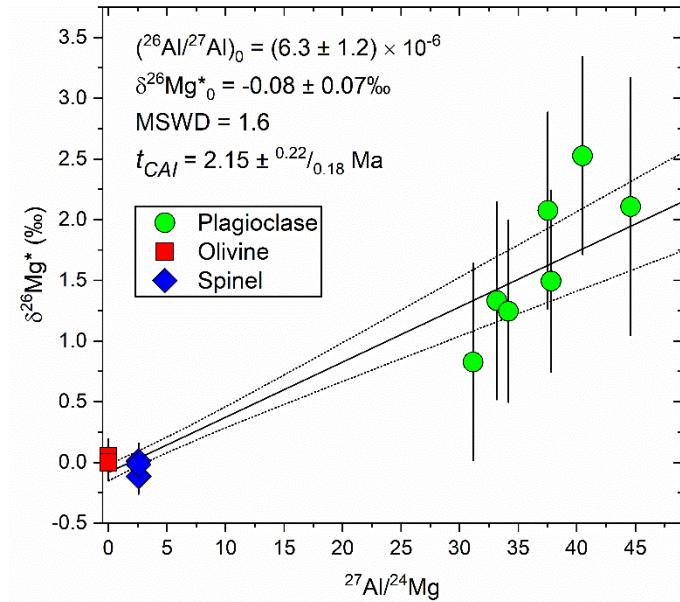
1320 Hf, Sc, Y, Ti, Sr, Ba, and REE) patterns with varying depletions of V. The reason for V depletion is
1321 unknown. Abundances of Cr, Mn, Rb decrease as their volatility increases. (b) Individual analysis of
1322 NWA 8276_A2. Trace element abundances of mottled mesostasis are almost identical to the clean
1323 glass. (c) Individual analysis of high-Ca pyroxene and mesostasis in MET 00526_A1. High-Ca
1324 pyroxene shows enrichments of Sc, Ti, V, and HREEs compared to its microcrystalline mesostasis.
1325 (d) Individual analysis of MET 00526_A2. An analysis of scapolite shows complementary REEs to
1326 microcrystalline mesostasis. In contrast, another scapolite analysis is almost identical to mesostasis,
1327 which might have sampled a significant amount of mesostasis beneath. (d) Bulk trace elements of
1328 OC ARCs in literature, in which Groups I, II, and III are plotted in different colors (MacPherson and
1329 Huss, 2005; Ebert and Bischoff, 2016). Abbreviations: MC Mes = micro-crystalline mesostasis;
1330 others are the same as in Fig. 1.

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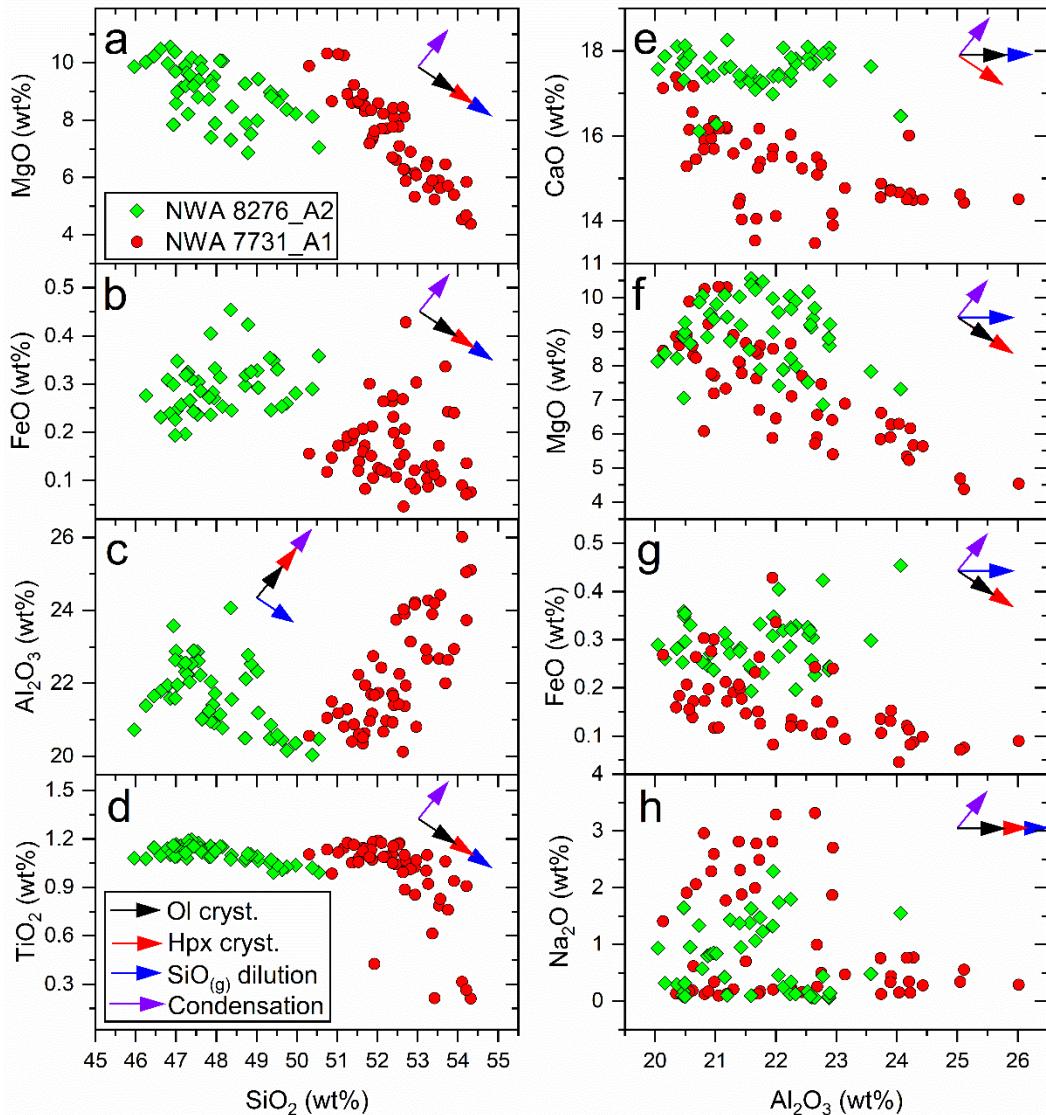
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1335 Fig. 6. Oxygen isotope ratios of major minerals in the six ARCs studied. MET 00526_A2 and
1336 NWA 8649_A1 display heterogeneity in the mass-independent fractionation of oxygen isotope
1337 (#), while the remaining four are homogeneous. Two analyses on the mottled regions of glassy
1338 mesostasis in NWA 8276_A2, which show ~5 ‰ $\delta^{18}\text{O}$ and ~1 ‰ $\Delta^{17}\text{O}$, are not plotted.
1339 Abbreviations: TF = terrestrial fractionation line; PCM = primitive chondrule mineral line
1340 (Ushikubo et al., 2012); others are the same as in Fig.1.
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1343 Fig. 7. The Al-Mg isochron diagram for the plagioclase-bearing ARC, MET 00526_A2. The solid
 1344 line represents the regression, and the dashed lines are 95 % confidence intervals on the fit.
 1345 Uncertainties for $(^{26}\text{Al}/^{27}\text{Al})_0$ and $(\delta^{26}\text{Mg}^*)_0$ are 95 % confidence levels.

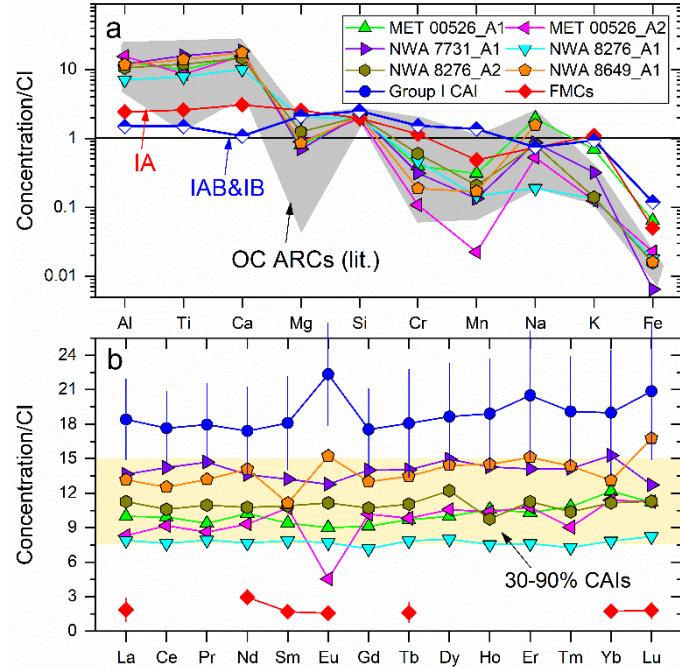
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1348 Fig. 8. Correlations of major elements in the glassy mesostasis of NWA 8276_A2 and NWA
 1349 7731_A1. The black and red arrows indicate the evolution trend of glassy mesostasis due to olivine
 1350 and high-Ca pyroxene crystallization, respectively. In contrast, the blue arrow indicates the dilution
 1351 effects on the concentrations of refractory elements like Al and Ti by the condensation of gaseous
 1352 SiO. The purple arrow indicates the evolution trends of gaseous element condensation.

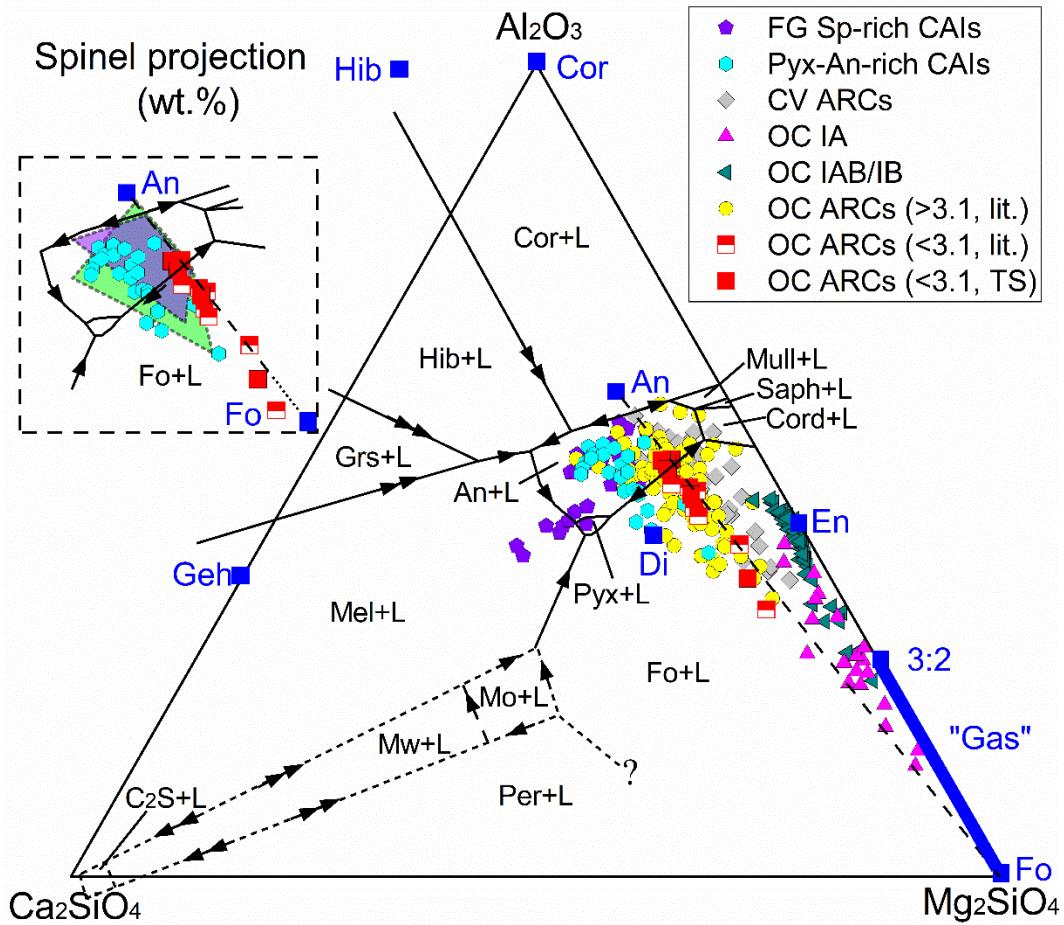
1353



1354

1355 Fig. 9. Calculated bulk major and REE concentrations of OC ARCs in this study. The ranges of REE
 1356 concentration in Group I CAIs and FMCs are both limited (Martin and Mason, 1974; Wänke et al.,
 1357 1974; Mason and Martin, 1977; Gooding et al., 1980; Grossman and Wasson, 1982, 1983; Mason
 1358 and Taylor, 1982; Kurat et al., 1984; Rubin and Pernicka, 1989; Swindle et al., 1991; Sylvester et al.,
 1359 1993; Huang et al., 2012; Davis et al., 2018; Torrano et al., 2019, 2023); thus, the portions of CAI
 1360 materials in the precursors of OC ARCs could be estimated using the CAI-FMC mixing model, which
 1361 are within the range of 30-90% (yellow region). The bulk major element compositions of OC ARCs
 1362 from the literature (Bischoff and Keil, 1984; Krot and Rubin, 1994; MacPherson and Huss, 2005;
 1363 Nagahara et al., 2008; Jiang et al., 2015; Ebert and Bischoff, 2016; Ross et al., 2017; Varela and
 1364 Zinner, 2018) are plotted for comparison.

1365



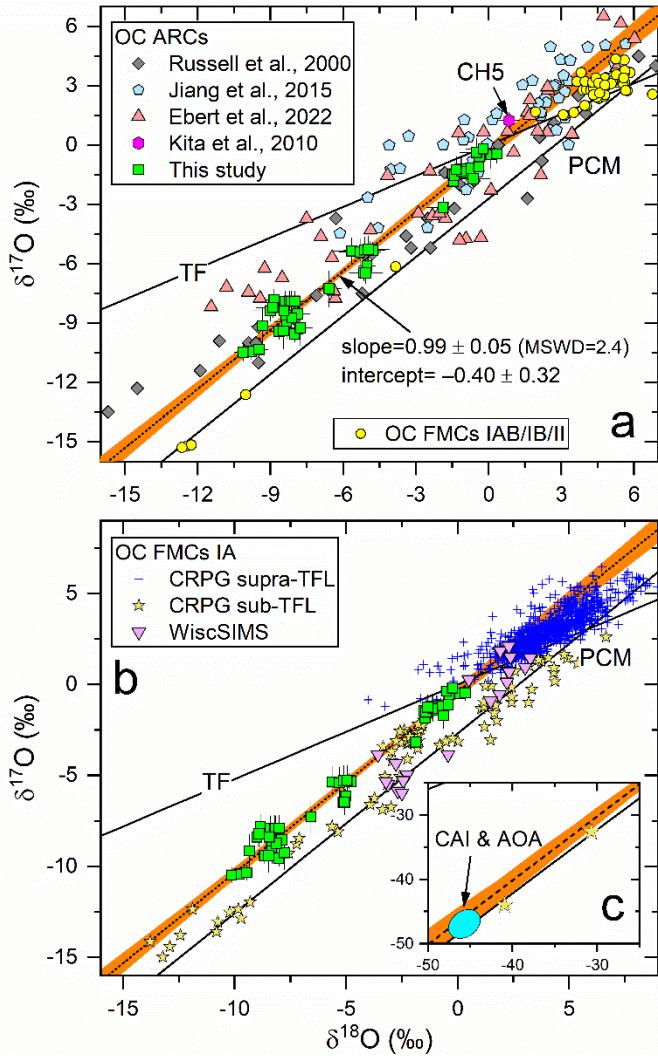
1366

1367 Fig. 10. Bulk compositions of OC ARCs, as plotted on the spinel-saturated ternary diagram of
 1368 Ca_2SiO_4 – Mg_2SiO_4 – Al_2O_3 . The phase diagram is adopted from Huss et al. (2001) and MacPherson
 1369 and Huss (2005). The data of OC ARCs with petrologic type ≤ 3.1 are from this study and literature
 1370 (Semarkona and NWA 7402; Rubin, 2004; MacPherson and Huss, 2005; Nagahara et al., 2008;
 1371 Ross et al., 2017). The data of OC ARCs with petrologic type > 3.1 are from Bischoff and Keil (1983),
 1372 Bischoff et al. (1989), Krot and Rubin (1994), MacPherson and Huss (2005), Jiang et al. (2015),
 1373 Ebert and Bischoff (2016), and Varela and Zinner (2018), with their bulk compositions corrected for
 1374 Fe–Mg and (N, K)–Ca exchanges. For comparison, literature data of CV ARCs (Sheng et al., 1991;
 1375 Zhang et al., 2020b), OC IA/IAB/IB chondrules (Jones and Scott, 1989; Jones, 1994; Tachibana et
 1376 al., 2003; Kita et al., 2010), and CV fine-grained spinel-rich (“FG Sp-rich”) CAIs (Krot et al., 2004b),
 1377 and pyroxene-anorthite-rich (“Pyx-An-rich”) CAIs in CV (namely type C) (Wark, 1987; Lin and Kimura,
 1378 1998), CO, and Acfer 094 chondrites (Krot et al., 2004a; Zhang et al., 2020a) are plotted. The blue

1379 segment on the Fo-Cor axis represents the composition range of ambient gas inferred from the bulk
1380 compositions trend of OC ARCs on the MgO-(CaO+Al₂O₃)-SiO₂ diagram (Fig. S4). The green and
1381 purple regions represent the bulk compositions of CAI components in the precursors of OC ARCs
1382 calculated by assuming two endmember mixing of CAIs and type I (IA&IAB/IB) chondrule materials
1383 or CAIs and the ambient gas, respectively (Table S10b). Abbreviations: Cor = corundum; Hib =
1384 hibonite; Grs = grossite; Geh = gehlenite; Mel = melilite; An = anorthite; Pyx= pyroxene; Di =
1385 diopside; En = enstatite; Fo= forsterite; Cord= corderite; Saph = sapphirine; Mull = mullite; Mo =
1386 monticellite; Mw = merwinite.

1387

1388

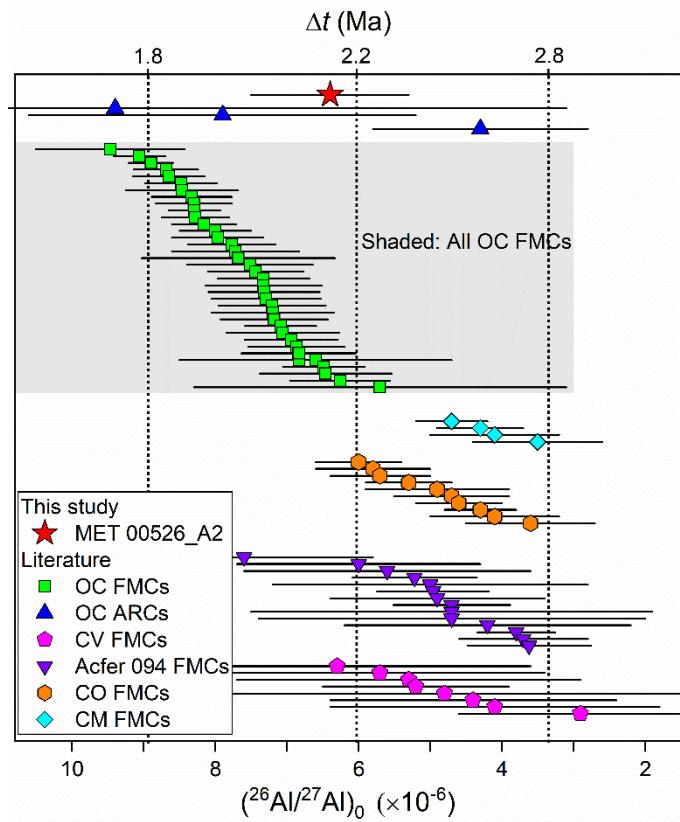


1389

1390 Fig. 11. The oxygen isotope mixing line of OC ARCs in this study. The equation of the mixing
 1391 line is $\delta^{17}\text{O} = (0.99 \pm 0.05) \times \delta^{18}\text{O} - (0.40 \pm 0.32)$ ($\text{MSWD} = 2.4$). The error envelope of the
 1392 mixing line is shaded in orange. In panel a, the oxygen isotope ratios of ARCs from
 1393 metamorphosed OCs (Russell et al., 2000; Jiang et al., 2015; Ebert et al., 2022) and OC type
 1394 IAB & B & II chondrules (Kita et al., 2010; Siron et al., 2021, 2022) are plotted for comparison. In
 1395 panel b, type IA chondrules analyzed by the CRPG-CNRS group (Piralla et al., 2021; Marrocchi
 1396 et al., 2024) and the WiscSIMS group (Kita et al., 2010; Siron et al., 2022) are shown in different
 1397 symbols; the former was displayed in two groups, i.e., supra-TFL (terrestrial fractionation line,
 1398 $\Delta^{17}\text{O} > 0\text{‰}$) and sub-TFL ($\Delta^{17}\text{O} < 0\text{‰}$), as suggested by Marrocchi et al., (2024). In panel c, the

1399 oxygen isotope region of CAIs and AOAs are indicated along with the two AOA-like relict
1400 olivines ($\Delta^{17}\text{O} < -15\text{\textperthousand}$) identified by Piralla et al., (2021), which are consistent with both the
1401 PCM and the OC ARC lines.

1402

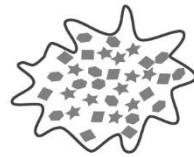


1403

1404 Fig. 12. Comparison of the Al-Mg age of MET 00526_A2 of this study to literature OC ARCs
 1405 (Russell et al., 1996; Russell et al., 1997) and FMCs in OCs (Siron et al., 2021, 2022), COs and
 1406 CMs (Fukuda et al., 2022), Acfer 094 (Ushikubo et al., 2013; Hertwig et al., 2019), and CVs
 1407 (Nagashima et al., 2017). A grey shaded area represents the range of $(^{26}\text{Al}/^{27}\text{Al})_0$ of OC
 1408 (L/LL<3.1) FMCs in earlier literature (Hutcheon and Hutchison, 1989; Kita et al., 2000;
 1409 Rudraswami and Goswami, 2007; Rudraswami et al., 2008; Villeneuve et al., 2009; Mishra et al.,
 1410 2010; Bolland et al., 2019; Pape et al., 2019). The significant older Al-Mg ages (~0.8 Ma) of two
 1411 OC FMCs determined from spinels by Piralla et al. (2023) were not plotted. If all OC FMC data
 1412 are considered, MET 00526_A2 formed within the period of OC chondrule-forming events. If
 1413 only OC FMC data from recent studies (Siron et al., 2021, 2022) were considered, MET
 1414 00526_A2 plots to their younger end, likely suggesting a late arrival of CAI materials from the
 1415 outer solar system into the OC chondrule-forming region.

1416

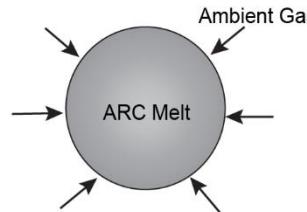
Stage I: Nebular condensation (~1300-1450 K)



Pyx-An-rich CAIs

- 1: ^{16}O -rich;
- 2: RLEs-rich (mostly flat REE);
- 3: No ^{50}Ti excess;
- 4: Indigenous or exotic in the inner disk;

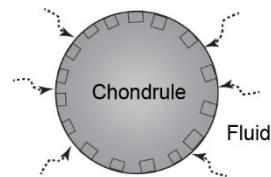
Stage II: Melting (>1673 K) and quenching (>1000 K/h to ~1 K/h)



Gas-melt interactions

- 1: Condensation of Mg, SiO, Fe, Na;
- 2: Condensed Mg:SiO~3:2 to 2:1 (atomic);
- 3: O exchange with ambient gas;

Stage III: Asteroidal modification (<873 K)



1: Bleaching (low pH, low-T)

Na, Ca, Al out and Fe, Ni, K in;
Glass dissolution;

2: Albitization (high pH, moderate-T)

Feldspathoids formation, Fe replaced Mg;
Became Na-ARCs after dehydration;

1417

1418 Fig. 13. Schematic diagram for the formation mechanism of OC ARCs. The cooling rates of OC
1419 ARCs are inferred from their textures, i.e., plagioclase-bearing (~10-500 K/h; Tronche et al.,
1420 2007; ~1 K/h; Wick and Jones, 2012), barred-olivine (100-7200 K/h; Lofgren and Lanier, 1990;
1421 Radomsky and Hewins, 1990; Tsuchiyama et al., 2004; or <10 K/h; Faure et al., 2022).
1422 Abbreviations: low-T: low temperature; high-T = high temperature; Na-ARCs = Na-Al-rich
1423 chondrules.

Table 1. Average major element compositions of constituent minerals determined by EPMA and the reconstructed bulk compositions of the six ARCs studied

ARC		MET 00526 A2 [#]					MET 00526 A1				NWA 7731 A1				
Size (μm)		430 × 250					570 × 300				440 × 440				
Minerals	Sp (5)	Ol (5)	Hpx (4)	Pl (11)	Bulk [‡]	Ol (12)	Hpx (19)	Mes (7)*	Bulk [‡]	Sp (1)	Ol (2)	Hpx (26)	Glass (53)	Bulk [‡]	
Vol%	3.2	13.6	16.7	42.1		7.7	16.1	76.2		0.0	0.1	37.5	62.3		
MgO	28.8	56.1	22.3	0.91	18.5	56.2	15.4	6.4	12.6	27.7	57.1	15.8	7.33	11.1	
SiO ₂	0.07	41.2	50.2	45.7	43.7	42.3	45.3	47.1	47.3	1.54	42.7	45.4	52.4	49.7	
Al ₂ O ₃	70.4	0.14	5.62	34.0	22.0	0.29	13.5	24.4	20.7	69.2	0.19	15.3	22.2	19.4	
CaO	0.03	0.36	17.4	18.7	13.9	0.42	21.6	14.8	15.1	0.74	0.34	20.7	15.3	17.7	
FeO	0.23	0.46	0.66	0.44	0.56	0.75	0.65	1.78	1.51	0.11	0.28	0.11	0.19	0.15	
Na ₂ O			0.01	0.60	0.30		0.05	1.84	1.37			0.03	1.04	0.61	
TiO ₂	0.44	0.15	3.02		0.85	0.13	1.98	0.45	0.72	0.41	0.13	1.40	0.98	1.16	
Cr ₂ O ₃	0.14	0.09	0.11		0.05	0.15	0.25	0.12	0.15	0.23	0.14	0.17	0.08	0.12	
V ₂ O ₃	0.15		0.12		0.05		0.08	0.03	0.04	0.31		0.09	0.06	0.07	
NiO		0.09	0.20		0.10	0.11	0.16	0.47	0.39		0.01	0.02	0.03	0.03	
MnO	0.01	0.05	0.03		0.01	0.08	0.06	0.08	0.08		0.05	0.02	0.04	0.03	
ZnO	0.06				0.00					0.05			0.00		
Total	100.3	98.7	99.6	100.4	100.0	100.4	99.1	97.5	100.0	100.3	100.9	99.2	99.7	100.0	
ARC		NWA 8276 A1					NWA 8276 A2				NWA 8649 A1				
Size (μm)		1100 × 700					580 × 580				200 × 200				
Minerals	Sp (6)	Ol (9)	Hpx (6)	Mes (10)*	Bulk [‡]	Ol (10)	Hpx (4)	Glass (49)	Bulk [‡]	Ol (2)	Hpx (11)	Glass (11)	Bulk [‡]		
Vol%	0.7	43.5	1.0	54.8		19.7	0.2	80.1		10.1	14.5	75.4			
MgO	28.9	57.2	18.5	10.1	32.6	57.2	14.4	8.95	19.8	56.5	14.4	6.32	13.5		
SiO ₂	0.08	42.6	48.6	47.3	44.8	42.6	45.4	48.1	47.1	41.4	44.9	49.4	48.3		
Al ₂ O ₃	70.7	0.23	8.7	20.7	11.4	0.23	14.6	21.7	17.0	0.08	16.2	22.1	18.9		
CaO	0.05	0.64	20.2	18.3	9.88	0.64	22.2	17.5	13.9	0.34	21.8	17.7	16.7		
FeO	0.48	0.47	0.37	0.38	0.42	0.47	0.37	0.34	0.37	0.32	0.34	0.39	0.38		
Na ₂ O			0.02	0.26	0.13		0.02	0.73	0.57		0.02	1.51	1.10		
TiO ₂	0.25	0.07	1.47	1.03	0.58	0.07	1.53	1.10	0.88	0.09	1.65	1.00	1.02		
Cr ₂ O ₃	0.23	0.12	0.32	0.23	0.18	0.12	0.49	0.26	0.23	0.05	0.10	0.07	0.07		
V ₂ O ₃	0.35		0.07	0.05	0.03		0.09	0.06	0.04		0.07	0.03	0.03		
NiO		0.05	0.05	0.04	0.04	0.05	0.04	0.09	0.08	0.04	0.04	0.07	0.06		
MnO	0.01	0.03	0.07	0.04	0.04	0.03	0.07	0.06	0.05	0.04	0.03	0.05	0.04		
ZnO	0.03				0.00				0.00				0.00		

Total	101.1	101.4	98.4	98.5	100.0	101.4	99.2	98.9	100.0	98.8	99.5	98.5	100.0
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#MET 00526_A2 contains 15.7 area% Na-Cl-bearing phases attributed to parent body alteration and has not been included in bulk composition calculation;

*Compositions of mesostasis were obtained using a defocused beam (15 μm). The numbers in brackets represent the number of analyses for each mineral.

Individual analysis result can be found in Table S2. \pm Bulk composition of a chondrule was recalculated from its modal mineral abundance (in wt%) and average mineral compositions; Abbreviations: Sp = spinel; OI = olivine; Hpx = high-Ca pyroxene; Pl = plagioclase; Mes = mesostasis.

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Table 2. Average trace element concentrations of constituent minerals determined by LA-ICP-MS and the reconstructed bulk trace element compositions of the six ARCs studied

ARCs	MET 00526 A1			MET 00526 A2			NWA 7731_A1	NWA82 76_A1	NWA82 76_A2	NWA86 49_A1	Group I CAI		OC FMCs	
	Hpx	Mes	Bulk	Na-rich	Mes	Bulk					Mean	SD	Mean	SD
N	2	3	5	4	4	8	6	6	23	3	33		165	
Zr	42.5	40.9	37.6	7.0	66.8	52.7	47.8	24.6	40.8	47.8				
Hf	1.6	1.1	1.1	0.2	1.9	1.5	1.4	0.6	1.2	1.4				
Sc	149	48.0	62.8	5.2	108	84.9	83.9	43.2	63.9	93.2				
Y	23.1	13.8	14.3	1.8	24.6	19.4	19.9	10.3	15.2	19.9				
Ti	11828	4208	5448	645	9661	7640	7185	3382	5492	6278				
Sr	8.7	83.7	62.2	71.2	39.8	31.4	97.7	51.3	74.1	114.6				
Ba	0.3	17.2	12.5	10.1	9.7	7.6	30.8	16.2	21.2	29.9				
V	240.0	29.3	66.6	22.4	17.2	18.1	178.3	67.3	99.9	25.3				
Cr	1863	1274	1279	82	787	635	1011	1057	1573	445				
Mn	448	875	743	90	229	183	370	383	495	212				
Rb	0.2	2.7	2.0	3.2	3.1	2.4	0.0	0.1	0.7	0.1				
La	1.0	2.8	2.2	0.5	3.3	2.6	3.2	1.7	2.5	3.0	4.2	0.7	0.44	0.24
Ce	3.6	7.3	6.0	1.6	9.7	7.6	8.8	4.4	6.3	7.7	10.8	1.6		
Pr	0.8	1.0	0.9	0.2	1.4	1.1	1.4	0.7	1.0	1.2	1.6	0.3		
Nd	4.7	5.2	4.6	0.9	7.2	5.7	6.2	3.2	4.7	6.3	7.4	1.4	1.35	0.31
Sm	2.0	1.4	1.4	0.1	2.6	2.1	1.9	1.0	1.5	1.6	2.5	0.5	0.25	0.08
Eu	0.0	0.6	0.5	0.6	0.4	0.3	0.7	0.4	0.6	0.8	1.2	0.2	0.09	0.03
Gd	2.5	1.8	1.8	0.6	3.4	2.7	2.8	1.3	2.0	2.5	3.2	0.4		
Tb	0.6	0.3	0.4	0.0	0.6	0.5	0.5	0.3	0.4	0.5	0.6	0.1	0.06	0.03
Dy	4.0	2.3	2.4	0.3	4.3	3.4	3.6	1.7	2.8	3.4	4.2	0.9		
Ho	0.9	0.6	0.6	0.1	1.0	0.8	0.8	0.4	0.5	0.8	1.0	0.2		
Er	2.7	1.6	1.7	0.2	3.0	2.3	2.3	1.1	1.8	2.4	3.1	0.7		
Tm	0.4	0.3	0.3	0.0	0.4	0.3	0.3	0.2	0.2	0.3	0.4	0.1		
Yb	3.1	2.0	2.0	0.2	3.2	2.5	2.5	1.2	1.7	2.1	2.9	0.7	0.28	0.10
Lu	0.4	0.3	0.3	0.0	0.5	0.4	0.3	0.2	0.3	0.4	0.7	0.5	0.05	0.07
CAI % ^a	40-75			30-75			40-100	25-60	40-80	50-80				
Low limit ^c	~80			~75			~80	~45	~65	~75				

^a: The portion of CAIs in the precursor of OC ARCs was estimated from their trace element concentrations using the “CAI-FMC hybrid” model and the “CAI + Gas” model (Table S10a). ^b: The low limit of the CAIs portion in the precursor of OC ARCs is estimated from their major element contents using the “CAI-FMC hybrid” and the “CAI-Gas” models (Table S10b). Individual analysis result can be found in Table S3. Data source for group I CAIs is: Martin and Mason (1974), Wänke et al. (1974), Mason and Martin (1977), Mason and Taylor (1982), Sylvester et al. (1993), Huang et al. (2012), Davis et al. (2018), and Torrano et al. (2019, 2023). Data source for OC FMCs is: Gooding et al. (1980), Grossman and Wasson (1982, 1983), Kurat et al. (1984), Rubin and Pernicka (1989), and Swindle et al. (1991).

Table 3. Oxygen isotope ratios of major minerals in the six ARCs studied

Chondrule	Texture	Spot	Mineral	$\delta^{18}\text{O}$	Unc.	$\delta^{17}\text{O}$	Unc.	$\Delta^{17}\text{O}$	Unc.	
<i>Heterogeneous chondrules</i>										
MET 00526_A2 [#]	Porphyritic	#1a	Sp	-9.8	0.4	-10.4	0.4	-5.3	0.3	
		#2a	Sp	-10.1	0.4	-10.5	0.4	-5.2	0.3	
		#3a	Ol	-8.7	0.4	-9.4	0.4	-4.9	0.3	
		#4a	Ol	-9.5	0.4	-10.3	0.4	-5.4	0.3	
		#6b	Ol	-9.3	0.5	-9.1	0.9	-4.3	0.9	
		#1b	Hpx	-8.3	0.5	-8.9	0.9	-4.6	0.9	
		#2b	Hpx	-8.5	0.5	-9.4	0.9	-5.0	0.9	
		#5b	Hpx	-8.3	0.5	-8.6	0.9	-4.3	0.9	
		#8b	Hpx	-7.9	0.5	-8.5	0.9	-4.4	0.9	
		#5a	Pl	-5.1	0.4	-6.5	0.4	-3.8	0.3	
		#6a	Pl	-5.0	0.4	-6.1	0.4	-3.5	0.3	
		#7a	Pl	-8.0	0.4	-9.6	0.4	-5.4	0.3	
		#3b	Pl	-7.8	0.5	-9.3	0.9	-5.2	0.9	
		#4b	Pl	-8.1	0.5	-8.7	0.9	-4.5	0.9	
		#7b	Pl	-6.6	0.5	-7.3	0.9	-3.9	0.9	
NWA 8649_A1	Glassy	#1	Gl	-0.6	0.7	-1.7	0.5	-1.4	0.3	
		#2	Gl	-5.1	0.7	-6.5	0.5	-3.8	0.3	
		#3	Gl	-1.9	0.7	-3.2	0.5	-2.2	0.3	
<i>Homogeneous chondrules</i>										
NWA 8276_A2 [*]	Glassy	#5	Gl (Altered)	4.2	0.7	2.7	0.7	0.5	0.4	
		#6	Gl (Altered)	4.7	0.7	3.3	0.7	0.9	0.4	
		Host (2Ol, 2Gl) [‡]		-0.2	0.5	-0.4	0.5	-0.3	0.3	
NWA 8276_A1	Glassy	Host (2Sp, 3Ol, 2Hpx, 3Gl) [‡]			-1.0	0.5	-1.3	0.4	-0.8	0.2
MET 00526_A1	Glassy	Host (2Ol, 4Hpx) [‡]			-5.1	0.6	-5.3	0.4	-2.7	0.2
NWA 7731_A1	Glassy	Host (3Hpx, 5Gl) [‡]			-8.5	0.5	-8.1	0.3	-3.6	0.2

[#]: MET 00526_A2 has been analyzed in the Jul. 2021 session (12 μm , "a") and the Sep. 2021 session (3 μm , "b");

^{*}: Two analyses on the mottled regions (likely altered) of NWA 8276_A2 have distinct oxygen isotope ratios compared to others on clean regions; [‡]: the number in the brackets represents the number of analyses for each mineral; Unc. = uncertainty, other abbreviations are the same as previous. Instrumental bias correction, data reduction, and data summary can be found in tables S4, S5, and S6, respectively.

Table 4. Al-Mg isotope data of MET 00526_A2

Mineral	No.	$^{27}\text{Al}/^{24}\text{Mg}$	2SE	$(\delta^{26}\text{Mg})^*$	2SE
Ol	#1	6.00E-03	1.07E-03	0.05	0.15
	#2	4.15E-03	7.37E-04	0.00	0.15
Sp	#1	2.57	2.62E-02	-0.11	0.15
	#2	2.58	2.63E-02	0.01	0.15
Pl	#3	2.57	2.62E-02	-0.02	0.15
	#1	33.2	0.42	1.33	0.81
	#2	31.1	0.39	0.83	0.81
	#3	37.5	0.49	2.08	0.81
	#4	44.6	0.54	2.11	1.06
	#6	34.1	0.40	1.25	0.75
	#8	37.8	0.46	1.49	0.75
	#9	40.5	0.52	2.53	0.81

abbreviations are the same as previous. Instrumental bias correction and data reduction details can be found in tables S7-S9.