Lithium- and oxygen-isotope compositions of chondrule constituents in the Allende meteorite

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Abstract

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We report in situ ion-microprobe analyses of Li- and O-isotope compositions for olivine, low-Ca pyroxene, high-Ca pyroxene, and chondrule mesostasis/plagioclase in nine chondrules from the Allende CV3 chondrite. Based on their mineralogy and O-isotope compositions, we infer that the chondrule mesostasis/plagioclase and ferroan olivine rims were extensively modified or formed during metasomatic alteration and metamorphism on the Allende parent asteroid. We exclude these minerals in order to determine the correlations between Li and both O and the chemical compositions of olivines and low-Ca pyroxenes in the chondrules and their igneous rims. Based on the O-isotope composition of the olivines, nine chondrules were divided into three groups. Average Δ^{17} O of olivines (Fo_{>65}) in group 1 and 2 chondrules are $-5.3 \pm 0.4\%$ and $-6.2 \pm 0.4\%$, respectively. Group 3 chondrules are characterized by the presence of 16 O-rich relict grains and the Δ^{17} O of their olivines range from -23.7 to -6.2%. In group 1 olivines, as Fa content increases, variation of δ^7 Li becomes smaller and δ^7 Li approaches the whole-rock value (2.4‰; Seitz et al., 2012), suggesting nearly complete Li-isotope equilibration. In group 2 and 3 olivines, variation of δ^7 Li is limited even with a significant range of Fa content. We conclude that Li-isotope compositions of olivine in group 1 chondrules were modified not by an asteroidal process but by an igneous-rim formation process, thus chondrule olivines retained Li-isotope compositions acquired in the protosolar nebula. In olivines of the group 3 chondrule PO-8, we observed a correlation between O and Li isotopes: In relict 16 O-rich olivine grains with Δ^{17} O of ~ -25 to -20%, δ^7 Li ranges from -23 to -3%; in olivine grains with Δ^{17} O > -20%, δ^7 Li is nearly constant ($-8 \pm 4\%$). Based on the Li-isotope composition of low-Ca pyroxenes, which formed from melt during the crystallization of host chondrules and igneous rims, the existence of a gaseous reservoir with a $\delta^7 \text{Li} \sim$ -11% is inferred.

Keywords: lithium, oxygen, chondrule, chondrite, asteroid, Allende, igneous rim, SIMS

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1. INTRODUCTION

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Type 3 carbonaceous and ordinary chondrites have long been considered as primordial materials, relatively unchanged since their formation; however, most have been altered by interaction with aqueous fluid on their parent asteroids (Krot et al., 1995; Zolensky et al., 2008; Brearley, 2014). Lithium is highly soluble in aqueous fluids (Brenan et al., 1998) and Li isotopes exhibit significant fractionation in geologic materials (δ^7 Li ranges from -20 to 40%) because of the large mass difference between ⁶Li and ⁷Li (e.g., Tomascak et al., 2016). Solid phases preferentially retain ⁶Li, while ⁷Li preferentially enters solution. These unique geochemical characteristics of Li make it possible to trace aqueous alteration processes on asteroids.

Despite the potential role of Li as a geochemical tracer, its behavior with respect to geological processes, operating in the early solar system, is not well understood. Hanon et al. (1999) reported a wide range of Li abundances in chondrules using secondary ion mass spectrometry (SIMS). Chaussidon and Robert (1998) found a significant range of Li-isotope compositions (δ' Li from -13 to +35%) in chondrules from the Semarkona LL3.01 (Kimura et al., 2008) chondrite and interpreted these variations as a result of heterogeneity inherited from chondrule precursors. McDonough et al. (2003) and Seitz et al. (2007) determined the Li-isotope compositions (δ^7 Li) of whole-rock carbonaceous and ordinary chondrites using inductively coupled plasma mass spectrometry (ICP-MS); the whole-rock δ^7 Li range from -3.5 to +3.9% (McDonough et al., 2003) and from +1.8 to +5.4% (Seitz et al., 2007), respectively. McDonough et al. (2003) found a negative correlation between δ^7 Li and the petrologic type of carbonaceous chondrites, suggesting that alteration on asteroids made the Li-isotope composition heavier. They inferred that isotopically heavy fluids had interacted with asteroidal materials to produce hydrous minerals that are enriched in ⁷Li. Maruyama et al. (2009) carried out in situ Li-isotope analyses of chondrule olivines in the Allende meteorite using SIMS and suggested that variation of $\sim 50\%$ observed in chondrule olivines could reflect a nebular process rather than an asteroidal process. Recently, Seitz et al. (2012) determined whole-rock Li-isotope compositions of chondrules, refractory inclusions, and dark inclusions from Allende and ordinary chondrites and demonstrated that the δ' Li in these materials ranges from -8.5 to +10%. Whilst these previous studies have contributed to our understanding of the behavior of Li in the early solar system, much concerning the role of asteroidal and nebular processes remains unclear.

Chondrules are the most abundant components in most chondrites and are believed to have formed in the solar nebula from clumps of precursor dust during repeatable transient heating events (e.g., Grossman, 1988; Jones, 1996a; Rubin, 1996; Hewins, 1997). Chondrules are composed of olivine, low-Ca pyroxenes, high-Ca pyroxenes, plagioclase, glass, metal, and sulfides. These phases respond differently to nebular and asteroidal processes. Variations of Li-isotope compositions observed in the above phases could have resulted from those of chondrule precursors, isotope fractionation

between silicates and melt, devolatilization, condensation, and interaction with isotope
 reservoirs, during nebular and asteroidal processes. Fully constraining the behavior of
 Li in these meteorites requires a more comprehensive approach involving analyses of a
 broader array of element and isotope compositions than previously investigated.

In order to understand the behavior of Li during nebular and asteroidal processes, we report the Li- and O-isotope compositions of individual minerals in the Allende chondrules and their igneous rims. We subsequently examine the formation and alteration processes associated with these phases.

2. EXPERIMENTAL METHODS

2.1. Major elements

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The major-element and minor-element compositions of mineral phases (olivine, low-Ca pyroxene, high-Ca pyroxene, and plagioclase) were determined by electron microprobe (EPMA) in wavelength-dispersive mode using a JEOL JXA-8530F. The analyses were conducted at routine conditions (15 kV acceleration voltage and 12 nA beam current) 87 and the oxide ZAF method was employed for matrix corrections. Calibration was under-88 taken using silicate, sulfide, oxide, and synthetic oxides minerals, which were assembled on a standard ASTIMEX, MINM25-53+FC mount (Astimex Standards Ltd., Canada). 90 Major-element compositions of glass were determined using a JEOL JSM-7001F SEM 91 equipped with energy dispersive X-ray spectrometers (EDS) and Oxford AZtec X-Max and X-act. The analyses were conducted under routine conditions (10 kV acceleration 93 voltage, 3 nA beam current, and 100 s integration times).

2.2. Oxygen-isotope compositions

Oxygen-isotope compositions were determined by SIMS using a modified Cameca ims-1270 at the Pheasant Memorial Laboratory (PML). Thick sections were coated with 75 nm of Au to avoid charging. At the same surface condition, isotope compositions were determined relative to that of a working standard, and matrix effects of minerals were estimated based on analyses of a working standard on a different mount. A Cs⁺ primary beam (200 pA) of 20 keV was focused and scanned on the sample surface by $5\times5~\mu\text{m}^2$, yielding a $6\times6~\mu\text{m}^2$ crater. Secondary O⁻ was accelerated to -10~kV and a normal incident electron gun was used to compensate positive charging on the sputtered area. In order to minimize the generation of OH⁻ secondary ions, a pressure of less than 5×10^{-9} Torr was maintained in the specimen chamber using a cold trap filled with liquid nitrogen. A contrast aperture of $400~\mu\text{m}$ was used and an optical gate of $1300\times1300~\mu\text{m}^2$, which corresponds to $25\times25~\mu\text{m}^2$ on the sample surface, was inserted and an energy band pass set from -10 to 30 eV. The mass-resolution was set to 5,000~for $^{17}\text{O}^-$ and 2,400~for $^{16}\text{O}^-$ and $^{18}\text{O}^-$. At these resolutions it was possible to

resolve OH⁻ interference at less than the 0.1% level. The pre-sputtering time, including auto peak-centering, was 190 s. Signal intensities were simultaneously determined by a Faraday cup mounted on trolley L2 ($^{16}O^-$) and pulse counting was done using electron multipliers located on axial ($^{17}O^-$) and trolley H1 ($^{18}O^-$). Typical ion intensities for $^{16}O^-$, $^{17}O^-$, and $^{18}O^-$ were 50M, 20k, and 100k cps, respectively. The ion-integration times were 14.5 s in each cycle, and each run consisted of 40 cycles. Typical external precision, estimated by repeated analyses of a working standard, was 0.3% (1SD) for both $\delta^{18}O$ and $\delta^{17}O$. The deviation of oxygen's isotope ratios from the terrestrial mass fractionation line is represented by $\Delta^{17}O = \delta^{17}O - 0.52 \times \delta^{18}O$ (Clayton, 1993).

Matrix effects on instrumental mass fractionation are known to occur for oxygenisotope measurements (Hervig et al., 1992; Eiler et al., 1997; Leshin et al., 1997; Gurenko et al., 2001; Gurenko and Chaussidon, 2002; Isa et al., 2017). In order to understand the matrix effects, we first attached importance to chemical varieties represented by $X_{\rm Mg} \ (\equiv \frac{[{\rm Mg}]^{\rm mol}}{[{\rm Fe}]^{\rm mol}+[{\rm Mg}]^{\rm mol}}), \ X_{\rm Ca} \ (\equiv \frac{[{\rm Ca}]^{\rm mol}}{[{\rm Fe}]^{\rm mol}+[{\rm Mg}]^{\rm mol}+[{\rm Ca}]^{\rm mol}}), \ {\rm and} \ X_{\rm Na} \ (\equiv \frac{[{\rm Na}]^{\rm mol}}{[{\rm Ca}]^{\rm mol}+[{\rm Na}]^{\rm mol}+[{\rm K}]^{\rm mol}}).$ We then examined the matrix effects through comparison with a working standard, San Carlos olivine $(\Delta\delta^{18}{\rm O}_{\rm ol\cdot sc}^{\rm phase})$, analyses of reference olivines $({\rm Fo}_{48-100})$, low-Ca pyroxenes $({\rm En}_{88-91}{\rm Wo}_{0.3-2})$, high-Ca pyroxenes $({\rm En}_{45-50}{\rm Wo}_{37-50})$, plagioclases $({\rm Ab}_{40-98})$, and glass $({\rm Ab}_{53-100})$ (Appendix 1.1. and Table S1). We found that the matrix effects were mass dependent. Thus we calculated the matrix effects for $\delta^{17}{\rm O}$ from that of $\delta^{18}{\rm O}$ $(\Delta\delta^{17}{\rm O}=0.52\times\Delta\delta^{18}{\rm O})$ and no correction was necessary $\Delta^{17}{\rm O}$.

In this study, we determined O-isotope compositions of olivine (Fo₆₂₋₁₀₀), low-Ca pyroxene (En₉₀₋₉₈Wo₁₋₉), high-Ca pyroxene (En₅₄₋₇₄Wo₂₆₋₄₅), and plagioclase (Ab₁₇₋₁₉) in Allende chondrules. The estimated matrix effects were negligibly small for olivine Fo₇₀₋₁₀₀, and plagioclase Ab₁₇₋₁₉, being within a reproducibility of ~0.2–0.3‰ (1SD) δ^{18} O for the working standard, thus we did not apply the matrix effect correction for these phases. Whereas for olivine (Fo₆₂₋₇₀), low-Ca pyroxene (En₉₀₋₉₈Wo₁₋₉), and high-Ca pyroxene (En₅₄₋₇₄Wo₂₆₋₄₅), we confirmed existence of significant matrix effects related with their chemical compositions (Table S2 and Figure S1). Therefore, we defined formula to represent their matrix effects, and corrected them for olivines and pyroxenes with particular chemical compositions (Appendix 1.1.).

2.3. Lithium concentrations and isotope compositions

Lithium-isotope compositions were determined by SIMS using the modified Cameca ims-1270 at the PML. Thick sections were coated with 30 nm of Au to avoid charging. O-isotope and Li-isotope measurements were carried out onto the same spots. However, in some cases spot locations differed by $20 \, \mu \text{m}$. The primary O⁻ ion beam was accelerated to $-23 \, \text{keV}$ and the primary beam current was varied from 1 to 15 nA to maintain constant secondary ion-intensity. A contrast aperture of $400 \, \mu \text{m}$ was used. An optical gate that corresponds to $50 \times 50 \, \mu \text{m}^2$ on sample surface was inserted and the energy band

pass was set to 0 ± 50 eV. In order to attain a mass resolution of 1200, which is sufficient to eliminate interference from $^6\text{LiH}^+$, the width of the entrance- and exit-slits was set to 400 and 1200 μm , respectively. The intensity ratio of $^6\text{Li}^+$ and $^7\text{Li}^+$ was determined in magnetic peak-jumping mode by ion counting with an electron multiplier. Integration times for $^6\text{Li}^+$ and $^7\text{Li}^+$ were 4 and 1 s in each cycle, respectively, and each run consisted of 110 cycles. Craters were less than 20 μm in diameter. Lithium concentrations for the analyzed spots were estimated using ($^7\text{Li}^+/\text{I}_{PRI}$), where I_{PRI} is the primary ion-probe current.

As was the case for O-isotope analysis, instrumental mass fractionation represents one of the major difficulties in high-precision Li-isotope analysis by SIMS (e.g., Bell et al., 2009). Isotope compositions were determined by the formula $\delta^7 \text{Li} = \delta^7 \text{Li}^{\oplus} - \Delta \delta^7 \text{Li}^{\text{phase}}$, where $\delta^7 \text{Li}^{\oplus}$ is the ion-intensity ratio that was used to correct the instrumental mass fractionation ($\delta^7 \text{Li}^{\oplus} \equiv \delta^7 \text{Li}^+ - f_{\text{IMF}}$), and $\Delta \delta^7 \text{Li}^{\text{phase}}_{\text{ws}}$ is the relative matrix effect of a particular phase relative to a working standard. Note that f_{IMF} is estimated using the working standard.

To evaluate the matrix effects relative to working standards, we analyzed reference olivine (Fo₁₋₉₉), low-Ca pyroxene (En₉₈Wo_{0.1}), high-Ca pyroxene (En₈₂₋₈₄Wo₃₁₋₃₉), and plagioclase (Ab₄₂₋₇₂) (Appendix 1.1. and Table S1). Significant matrix effects were recognized for olivines with Fo₁₋₉₉ and between the working standards and the other reference materials. Therefor, we estimated $\Delta \delta^7 \text{Li}_{ws}^{\text{phase}}$ for olivine, pyroxene, and plagioclase based on linear correlations between the matrix-related bias and the chemical composition of those phases (Table S3 and Figure S2). For olivine (Fo₆₂₋₁₀₀) in the Allende chondrules, matrix effects were estimated based on a linear correlation with chemical varieties, and the matrix effects were corrected. The estimated matrix effects of low-Ca and high-Ca pyroxenes (Wo₁₋₉ and Wo₂₆₋₄₅) and plagioclase (Ab₁₇₋₁₉) in chondrules relative to the working standard en-sl1 were negligibly small, being within or comparable to analytical reproducibilities (1SD) for the working standards, en-sl1 (δ^7 Li 2.6‰) or ol-sc1 (δ^7 Li 0.6‰). Thus we did not apply the matrix effect correction on the pyroxenes and the plagioclase; further details are described in Appendix 1.1.

7 3. RESULTS

3.1. Sample Descriptions

Nine chondrules, including chondrule fragments, were hand-picked after disaggregation from the Allende meteorite and embedded in a resin mount with working standard materials (San Carlos olivine). They are shown in Figure 1 and classified as porphyriticolivine (PO) chondrules, barred-olivine (BO) chondrules, and a barred-olivine-pyroxene (BOP) chondrule (Scott and Krot, 2014; Jones, 1990, 1994, 1996b,a). The major-element compositions for olivine, low-Ca pyroxene, high-Ca pyroxene, and plagioclase

in the chondrules are summarized in Table 1.

BOP-2 is a 2.0 mm-sized compound chondrule (Figs. 1a, 2a, 2b, S3a), which consists of a 1.0 mm-sized inner BO, a 2.0-mm sized outer BOP, and an igneous rim. The inner BO consists of olivine (Fo_{88-99}) , mesostasis, and opaques. Olivine bars are typically 50 μ m in width, elongated along a north east direction and surrounded by a 100 μ m thick olivine and opaque rich shell. The mesostasis is dominated by An-rich plagioclase (Ab₁₈₋₂₄An₇₆₋₈₂), low-Ca pyroxene (En₉₄Wo₅), high-Ca pyroxene $(En_{60-78}Wo_{21-39})$, Ab-rich glass $(Ab_{84-89}An_{5-11})$, and opaques. The outer BOP consists of olivine (Fo₉₅₋₉₉), mesostasis, low-Ca pyroxene (En₉₀₋₉₉Wo₉₋₁), high-Ca pyroxene $(En_{57-74}Wo_{42-26})$, and opaques. The typical width of olivine and low-Ca pyroxene bars is 100 and 200 μ m, respectively. Some barred olivines are surrounded by low-Ca pyroxenes. The mesostasis consists of Ab-rich glass (Ab_{81–88}An_{19–5}), low-Ca pyroxene $(En_{91-93}Wo_{7-6})$, high-Ca pyroxene $(En_{60-71}Wo_{39-28})$, and opaques. Bars of olivine and low-Ca pyroxene, elongated along a north east direction, are surrounded by a shell with a thickness of 140 μ m. The shell consists of olivine and opaques, and is surrounded by an igneous rim of low-Ca pyroxene, olivine, and opaques. Olivine is anhedral and enclosed by low-Ca pyroxene (Fig. 2a).

PO-6 is a 2.3 mm-sized fragment of a PO chondrule with an igneous rim (Fig. 1b). The main region consists of olivine (Fo₆₃₋₉₉) and mesostasis, and the igneous rim consists of olivine, low-Ca pyroxene (En₉₆₋₉₈Wo₁), high-Ca pyroxene (En₅₈₋₆₆Wo₄₀₋₃₂), and opaques. The chondrule mesostasis consists of albitic glass (Ab₈₈₋₈₉An₅) and high-Ca pyroxene (En₇₀₋₈₀Wo₁₇₋₂₆). Typical sizes of olivine in the chondrule and its igneous rim are $100 \,\mu\text{m}$ and $20 \,\mu\text{m}$, respectively.

BO-4 is a 1.7 mm-sized BO chondrule composed of olivine (Fo_{73–99}), mesostasis and opaques and surrounded by a 100 μ m thick olivine (Fo_{90–95}) shell of (Figs. 1c, S3b). Olivines in the main region occur as bars aligned at two angles. Typical bar width is 20 μ m, and the largest one is 200 μ m (with a length of 800 μ m). The mesostasis consists of An-rich glass (Ab₉An₉₁), anhedral low-Ca pyroxene (En₉₂Wo₇), anhedral high-Ca pyroxene (En_{56–89}Wo_{11–37}), and opaques.

PO-7 is a 1.3 mm-sized fragment of a PO chondrule (Figs. 1d, 2c) with an igneous rim. The main region consists of olivine (Fo₉₀₋₉₈) and albitic mesostasis (Ab₈₇An₆). Whereas the the igneous rim is composed of olivine, low-Ca pyroxene (En₉₈Wo₉), and opaques.

PO-1 is a 1.7 mm-sized fragment of a PO chondrule (Fig. 1e). The chondrule consists of olivine (Fo₆₅₋₁₀₀), euhedral low-Ca (En₉₂₋₉₅Wo₆₋₄) and high-Ca (En₅₄₋₅₉Wo₄₅₋₄₀) pyroxene and mesostasis (Fig. 1e). The mesostasis is dominated by Ab-rich glass (Ab₆₃₋₇₃An₃₂₋₂₁) with anhedral high-Ca pyroxene (En₂₄₋₄₇Wo₃₄₋₄₉). The typical size of olivine crystals is 80 μ m. Olivines near the exterior of the chondrule are enriched in FeO compared to those in its core (Fo₈₂₋₉₆ vs Fo₈₆₋₉₉).

PO-3c is a 160 μ m-sized fragment of a PO chondrule that consists of olivine (Fo₆₂₋₉₃)

(Fig. 1f). Similarly, PO-3s is a 180 μ m-sized fragment of a PO chondrule that consists of olivine (Fo_{86–99}) (Fig. 1g).

PO-8 is a 1.6 mm-sized PO chondrule surrounded by an igneous rim (Figs. 1h, 2d, S3c). The chondrule has a core-mantle structure. The diameter of the core region is 500 μ m and the thickness of the mantle and igneous rims are 500 μ m and 100 μ m, respectively. All three units are dominated by olivine (Fo₈₁₋₉₈) and opaques and their typical size in the mantle region is 40 μ m. In addition to the aforementioned phases, the mantle region contains mesostasis, whilst the igneous rim comprises of low-Ca (En₉₂₋₉₃Wo₆₋₅) and high-Ca pyroxene (En₅₇₋₆₆Wo₄₂₋₃₃). The mesostasis consists of An-rich plagioclase (Ab₁₇₋₁₉An₈₃₋₈₁).

PO-3n is a 600 μ m-sized fragment of a PO chondrule, which is composed of olivine (Fo_{77–95}), mesostasis, anhedral low-Ca pyroxene (En_{96–98}Wo_{2–1}), and opaques (Fig. 1i). The mesostasis consists of anorthitic glass (Ab_{8–9}An_{92–91}). The typical size of the olivine is 50 μ m. This chondrule is surrounded by fayalitic olivines (shown in the lower left of Fig. 1i).

3.2. Oxygen-isotope compositions

All the chondrules described above were measured for their oxygen-isotope compositions and these analyses are shown in Table S4 and Figure 3. The isotope compositions of minerals and the chondrule they were measured in are summarized in Table 2. On the O isotope diagram shown in Figure 3, δ^{18} O and δ^{17} O values of chondrule minerals (olivine, low-Ca pyroxene, high-Ca pyroxene, and plagioclase) scatter around the PCM (primitive chondrule mineral) line (slope of 0.987 and intercept of -2.70; Ushikubo et al., 2012) and the Y&R line (slope of 1.00 ± 0.03 and intercept of -1.04; Young and Russell, 1998). These O-isotope compositions are in good agreement with those previously obtained by whole rock and in situ analyses of CV chondrules (Clayton, 1981; Rubin et al., 1990; Jones et al., 2000; Rudraswami et al., 2011).

Based on the O-isotope composition of the olivines, the nine chondrules studied here were divided into three groups. Average $\Delta^{17}O$ values of olivine (Fo_{>65}) in chondrules BOP-2, PO-6, and BO-4 (group 1) are -5.3, -5.2, and -5.4%, varying by <1% (1SD). Average $\Delta^{17}O$ values of olivine (Fo_{>65}) in chondrules PO-7, PO-1, PO-3c, and PO-3s (group 2) are -6.1, -6.5, -6.3, and -6.0%, varying by <1% (1SD). The average $\Delta^{17}O$ of olivine compositions in chondrules of group 1 and 2 are $-5.3\pm0.4\%$ and $-6.2\pm0.4\%$, respectively. Olivines in chondrules PO-8 and PO-3n (group 3) show variations in $\Delta^{17}O$ from -23.7 to -6.2%.

In group 1, the δ^{18} O of olivine grains range from δ^{18} O -6% to -3%, pyroxene is similar but slightly more δ^{18} O-rich, and plagioclase in BOP-2 and FeO-rich olivines in PO-6, are a lot more δ^{18} O-rich. In group 2, the δ^{18} O of olivine, low-Ca pyroxene, and high-Ca pyroxenes are $\sim -6\%$, and FeO-rich olivines in PO-1 and PO-3c are a lot more δ^{18} O-rich. In group 3, δ^{18} O of olivines range from -50% to -10%. O-isotope

compositions of chondrule constituents in each group are summarized in Table 3 and Figures 5a,c,e. Further details are described in Appendix 1.2.

3.3. Lithium concentrations and isotope compositions

All chondrules measured for O-isotope compositions were also measured for Li-isotope compositions. All analyses are shown in Table S4 and Figure 4.

Low-Ca pyroxenes and plagioclases are the most Li enriched and depleted phases, respectively. Li-isotope compositions range from $\delta^7 \text{Li}$ –33‰ (low-Ca pyroxene) to +39‰ (olivine). There is no obvious correlation between Li concentration [Li] and isotope composition (Fig. 4). Li concentration and isotope compositions of minerals that are organized by occurrences in each chondrule are summarized in Table 2. The portion of results is visualized in Figure 2. Spatial distributions of [Li] and $\delta^7 \text{Li}$ are not systematic (Fig. 2).

The Li-isotope composition of chondrule constituents in each group is summarized in Table 3 and Figures 5b,d,f. Further details are described in Appendix 1.3.

4. DISCUSSION

4.1. The origin of phases in Allende chondrules

The Allende CV3 meteorite consists of chondrules, refractory inclusions and matrix and experienced significant alteration (e.g., Krot et al., 1995). Prior to investigation of Li behavior, we will evaluate the petrogenesis and alteration of Allende chondrule constituents.

Olivine occurs as phenocrysts and relict grains in chondrules (Ol) and igneous rims (Ol^{irim}), and as FeO-rich rims around phenocrysts (Ol^{FeO}). Chondrule olivine phenocrysts are typically euhedral, while olivine within igneous rims is anhedral. Some olivines are MgO-rich (Fo $_{\geq 95}$), whereas others possess higher Fe/Mg ratios (Fo $_{65-95}$). Igneous rims are present in about 50% of Allende chondrules and are also referred to as coarse-grained rim. Igneous rims are mostly composed of olivine and low-Ca pyroxene, although sulfide and metallic Fe-Ni are also present in minor amounts (Rubin, 1984). On average, Ol^{irim} is more enriched in Fe (Fo $_{78-92}$) relative to Ol. Ol^{FeO} is a minor constituent which occurs adjacent to Ol and forms euhedral crystals with significant Fe contents (Fo $_{\leq 65}$). Fe-enriched olivine is widespread in oxidized CV chondrites, being found in the rim, along cracks, and around Fe, Ni metal and sulfide inclusions, which form on forsteritic olivine phenocrysts (Brearley, 2014). The Fe-enriched olivines are formed through alteration on the parent body (Krot et al., 1995, 1998).

For Allende, Acfer 094, and CO chondrites, Δ^{17} O values of chondrules display a bimodal distribution with peaks at -5 and -2.5% (Rudraswami et al., 2011; Ushikubo et al., 2012; Tenner et al., 2013, 2015). Rudraswami et al. (2011) demonstrated that

O-isotope compositions of Allende chondrules formed from melt with a former Δ^{17} O reservoir range of -6 to -4.5%. Similarly, O-isotope compositions of Ol, in group 1 and 2 chondrules from this study, suggest olivine formed from melt with a former O-isotope reservoir during chondrule formation, as they cooled from their peak temperature.

Chondrules PO-8 and PO-3n show internal O isotope variation above analytical uncertainties. The variations observed within the two chondrules are similar to those of heterogeneous chondrules from Mokoia, Allende, and Acfer 094 (Jones et al., 2004; Rudraswami et al., 2011; Ushikubo et al., 2012). The two chondrules contain olivine that is significantly ¹⁶O-rich, which likely represents relict material that formed prior to chondrule formation, such as related to refractory inclusions and amoeboid olivine aggregates.

Group 1 and 2 chondrules were either more extensively melted or did not contain such relict grains. Chondrule BOP-2 consists of five layers (inner BO, shell of inner BO, outer BOP, shell of outer BOP, and igneous rim), and olivines from the five layers are indistinguishable within analytical uncertainties, suggesting that efficient O isotope exchange occurred between ambient gas and chondrule melt.

Plagioclase (Pl) is the last silicate to crystallized from a chondrule melt and occurs in the main chondrule as a constituent of the mesostasis. Pl has an average $\Delta^{17}O$ of $-3.3\pm0.5\%$ and is significantly depleted in ^{16}O compared to Ol, which is rather consistent with the average $\Delta^{17}O$ of the matrix and whole rock (-2.87 and -3.52%; Clayton and Mayeda, 1999). During parent body metamorphism, the O-isotope composition of $10~\mu$ m-sized anorthitic plagioclase could be completely modified, when considering the experimentally determined self-diffusion rate (Ryerson and McKeegan, 1994; Rudraswami et al., 2011). Pl can originate from either a chondrule formation or asteroidal process. Even in the case of chondrule formation, the ^{16}O -poor composition must result from an asteroidal process.

Ol^{FeO} is $10 \, \mu m$ in size, enriched in Fe (Fo₆₁₋₆₅) and infills $100 \, \mu m$ -sized Ol (Fig. 1f). Self-diffusion rates of O in olivine and pyroxene are several orders magnitude lower than that in anorthitic plagioclase (Ryerson and McKeegan, 1994; Cole and Chakraborty, 2001), so that O-isotope compositions recorded in these minerals during chondrule formation would not be affected by the metamorphism (Rudraswami et al., 2011). Ol^{FeO} has a similar O-isotope composition ($\Delta^{17}O$ –2.6 ± 0.9‰) to that of Pl and thus it is considered to be indicative of an asteroidal process (Imai and Yurimoto, 2003).

Low-Ca and high-Ca pyroxenes occur in both the main body of the chondrules and their igneous rims and are referred to as LPx and LPx^{irim} and HPx and HPx^{irim}, respectively. The O-isotope compositions of Ol, LPx, LPx^{irim}, and HPx of group 2 are indistinguishable. This finding suggests that the chondrule constituents formed from a melt with a homogeneous O-isotope composition, which reflects that of the ambient gas present during high temperature chondrule formation (Rudraswami et al., 2011).

Similarly to group 2, the $\Delta^{17}O$ values of group 1 Ol, Ol^{irim} , LPx, LPx^{irim} , and HPx

are essentially indistinguishable: -5.3 ± 0.4 , -5.0 ± 0.4 , -5.4 ± 0.4 , -5.1 ± 0.4 , and $-5.6 \pm 0.5\%$, respectively. However, δ^{18} O values for LPx, LPx^{irim}, and HPx (-3.1 ± 0.7 , -2.4 ± 1.1 , and $-2.5 \pm 0.9\%$) are higher than those for Ol ($-4.8 \pm 0.8\%$).

The existence of a gap in $\delta^{18}O$ between olivines and pyroxenes is consistent with the model of Chaussidon et al. (2008). They proposed that most of the olivine is relict and that the pyroxene was formed by interaction between the melt of a precursor dominated by olivine and gas of a constant isotope composition ($\delta^{18}O$, $\delta^{17}O = 1.2\%$, 0.6%, respectively). Using the proposed formulas by Chaussidon et al. (2008), the expected $\delta^{18}O$ and $\delta^{17}O$ of pyroxenes for group 1 is calculated to be -2.2% and -5.7%, respectively and our measured values for pyroxenes from group 1 chondrules are -3.1% and -7.0%, respectively. The observed $\delta^{18}O$ is 1% lighter than predicted and our observation is not consistent with the proposed gas having a constant O-isotope composition. Furthermore, the model of Chaussidon et al. (2008) cannot explain the similar $\Delta^{17}O$ of olivine and low-Ca pyroxene in the chondrules of group 1 and 2.

The O-isotope composition of olivine and pyroxene in group 1 and 2 chondrules have the same $\Delta^{17}O$ but the pyroxenes have heavier $\delta^{18}O$ than the olivines, which is opposite to the trend reported by Rudraswami et al. (2011), Ushikubo et al. (2012), and Tenner et al. (2013, 2015). These authors explain their observed trend by preferential evaporation of light isotopes from the melt during chondrule formation, prior to olivine crystallization. After crystallization of olivine, recondensing ambient gas could have supplied the melt with light isotope as it cooled, prior to pyroxene crystallization. However, our O-isotope compositions, for melt associated with group 1 chondrules, because heavier between the crystallization of Ol and LPx. Such a relationship could arise if evaporation was not terminated during crystallization of pyroxenes. Alternatively, meltgas reactions involving gas of a constant or changing O-isotope composition may have started to play a role during the crystallization of pyroxene.

Although the average Fe/Mg ratio of Ol^{irim} is larger than that of Ol, both the $\Delta^{17}O$ and $\delta^{18}O$ values of Ol^{irim} are indistinguishable to those of Ol. One possible explanation is that Ol^{irim} and LPx^{irim} formed during the later stages of chondrule formation from a melt with a fluctuating temperature and O-isotope composition. The addition of SiO to the outer part of a chondrule would also be necessary (Jones et al., 2005). However, the petrographic observation (Ol^{irim} is anhedral and poikilitically enclosed by LPx^{irim}) suggests that the igneous rim underwent partial melting, which was caused by a secondary heating event, after the formation of the chondrule (Krot and Wasson, 1995). We refer to the heating and cooling processes that formed LPx^{irim} from the melt as igneous-rim formation processes. The O-isotope compositions suggest that Ol^{irim} formed from melt during chondrule formation, with the Fe/Mg ratio modified and O-isotope composition maintained during the igneous-rim formation. Subsequently, LPx^{irim} formed from melt during the igneous-rim formation with the same O-isotope composition as LPx.

Rubin et al. (1990) determined O-isotope compositions of Allende main chondrule

bodies and their igneous rims using a conventional method and demonstrated that igneous rims tend to have higher $\Delta^{17}O$, up to 1.3% relative the main region. It is likely that the presence of secondary phases in the igneous rim, such as plagioclase, results in these higher $\Delta^{17}O$ values.

4.2. Lithium isotope systematics

Lithium concentration [Li] and δ^7 Li of whole-rock Allende are 1.93 μ g · g⁻¹ and 2.4% (Seitz et al., 2012), respectively. Lithium concentrations of olivine, mesostasis, and low-Ca pyroxene in Allende chondrules range from 0.1 to 3.5 μ g · g⁻¹. The δ^7 Li of the olivine in Allende ranges from -32 to +21% (Maruyama et al., 2009).

4.2.1 Effects of alteration processes on the CV asteroid and in the solar nebula

Relative to Ol and Ol^{irim}, Ol^{FeO} has a low Liconcentration (0.05 < [Li] < 0.3 μ g · g⁻¹) and its Li-isotope composition (δ^7 Li=3 ± 2‰) is consistent with that of the whole rock (Figs. 5d,e,f). The same feature is observed for Pl (0.008 < [Li] < 0.1 μ g · g⁻¹, δ^7 Li 0 ± 5‰). The identical Δ^{17} O of Ol^{FeO} and Pl and their common Li concentrations and isotope compositions, suggest that the Li signature of the two phases originated from the asteroidal reservoir. By assuming that Ol^{FeO} and Pl were equilibrated with the fluid, we can estimate the Li concentration of the fluid reservoir. Although Allende a experienced peak metamorphic temperature of ~ 550-600°C, aqueous/hydrothermal alteration occurred at a lower temperature (<300°C) (Krot et al., 1998). Caciagli et al. (2011) examined Li-partitioning between olivine and hydrous fluid, and plagioclase and hydrous fluid at 800–1000°C, which increases with decreasing temperature and plagioclase Ancontent. Using their partition coefficients for olivine and fluid 210⁺⁷²⁰₋₁₆₀, and for plagioclase (An₈₂Ab₁₈) and fluid 5.2^{+4.7}_{-2.5}, both at 300°C, we estimate the Li concentration of the fluid reservoir to be [Li] ~ 1⁺³₋₁ ng · g⁻¹ and ~ 8⁺⁷₋₄ ng · g⁻¹, which was calculated from a current [Li] of Ol^{FeO} (0.2 μ g · g⁻¹) and Pl (0.04 μ g · g⁻¹).

Asteroidal processes altered the concentrations and isotope compositions of mineral phases toward equilibrium. Once equilibrated, the Li concentrations and isotope compositions of a specific mineral would be identical regardless of its origin. However, a significant difference in Li distribution is observed between Ol and Ol^{irim} and LPx and LPx^{irim}, of different origins. This observation precludes Li equilibrium in olivine and low-Ca pyroxene during asteroidal processing, even though alteration and metamorphism were significant on the Allende parent body (Krot et al., 1998).

We examined Li behavior in group 1 olivines. The area of [Li]-and- δ^7 Li clusters in Figure 5b becomes smaller in the order of $Ol_{Fo>95}$, $Ol_{Fo<95}$, and Ol^{irim} , with constant averages of δ^7 Li. In Figure 6a, δ^7 Li is shown as a function of Fa content. Variation of δ^7 Li becomes smaller as Fe content increases, which suggests that the Li-isotope composition was moving towards equilibrium values as the Fe/Mg ratio increased. Bell et al.

(2008) also reported that variation of δ^7 Li is less in fayalitic olivines than in forsteritic olivines from Allende chondrules.

The ranges in the Fe/Mg ratio of group 2 and 3 olivines (from Fo₈₁ to Fo₁₀₀, and from Fo₇₇ to Fo₉₈) are similar to that of group 1 olivines (from Fo₇₈ to Fo₉₉). However, the trend, in which Li-isotope composition reaches equilibrium as the Fe/Mg ratio increases, is not observed in either group 2 or 3 olivines. If the Li-isotope equilibration observed for group 1 olivines occurred during asteroidal alteration, the Li-isotope compositions of group 2 and 3 olivines should have reached equilibrium as well. The Li-isotope composition could approach equilibrium through solid-state diffusion on the asteroid. Average Li concentration of Ol is higher than that of Ol^{FeO}. Therefore, Li of Ol was not equilibrated in the asteroidal environment. Since diffusion velocity of ⁶Li is faster than ⁷Li, alteration by asteroidal processes should give a negative-correlation between Li concentrations and isotope compositions. We do not see such a correlation and thus infer that modification of Li in olivines by a diffusion process was insignificant. Since Ol^{irim} of group 1 contains the highest Fe/Mg ratio and its δ ⁷Li is the most equilibrated (Fig. 5a), the Li-isotope equilibration of group 1 olivines must have taken place during igneous-rim formation.

4.2.2 Effects of igneous processes in the solar nebula

We find heavy Li ($\delta^7 \text{Li} \sim +40\%$) in both Ol and LPx in group 1 chondrules (Fig. 4a). Since the diffusion velocity of Li is faster than O both in solid materials and melt (Lowry et al., 1981; Wendlandt, 1991; Dohmen et al., 2002; Parkinson et al., 2007; Dohmen et al., 2010), the Li isotopes of the melt must have been homogenized at the same time as the O isotopes. The variation of δ^7 Li in Ol of group 1 should have been obtained after homogenization of the melt. Since the isotope fractionation between silicate and melt within the temperature range of chondrule formation is negligible, devolatilization is the only process that can give rise to the variation in isotope composition in the melt. In case of an evaporative loss of Li, the Li remaining in the melt would be enriched in the heavy isotope (Chaussidon and Robert, 1998). A variation of Li-isotope composition by $\sim 50\%$ is observed in Ol_{Fo>95}, which is the least Li altered phase during igneousrim formation. Since devolatilization makes the melt heavier and the lightest Li-isotope composition of $Ol_{Fo>95}$ in group is $\delta^7 Li \sim -10\%$, we infer the Li-isotope composition of the melt during homogenization of the O-isotope composition should have been δ^7 Li $\sim -10\%$. We propose that the Li-isotope composition of the melt evolved from $\delta^7 \text{Li} \sim$ -10% to +40%.

The abundance of Na remained relatively constant during chondrule formation (Alexander et al., 2008). Prevention of the evaporation of Na may require that chondrules formed in regions with high dust/gas ratios (Alexander et al., 2008). Fractionation of K isotopes could have been avoided by gas-melt isotope exchange during chondrule formation, if the K-vapor pressure was significantly high (Alexander et al., 2000). Since

Li is a trace element, pressure of Li-vapor was not as high as those of Na-vapor and K-vapor. If the Li-isotope fractionation observed by this study contributed to Rayleightype evaporation, >90% of Li in chondrule melt would have been transported into gas, with an isotope fractionation factor $\alpha = (^7\text{Li}/^6\text{Li})_{\text{vapor}}/(^7\text{Li}/^6\text{Li})_{\text{melt}} \sim 0.993$ (Kasemann et al., 2005).

The melt on LPx formation had different O and Li isotopes relative to the early melt. An obvious negative-correlation between concentration and isotope composition is not observed in $Ol_{Fo>95}$ (Figs. 4a-c, 5b). However, we infer that the variation in $\delta^7 Li$ of the melt, produced by devolatilization, is maintained in or recorded by the olivines, but the gas-melt Li-isotope exchange during igneous rim formation obscured the negative-correlation.

The variation in both Li-isotope composition and Fe/Mg ratios in group 1 olivine and the preservation of variation in Li-isotope composition, but modification of Fe/Mg ratio in group 2 and 3 olivines, is consistent with the conclusion of Parkinson et al. (2007). They state that diffuses 4–8 times slower than Fe-Mg in olivine and together with our findings contradicts the report by Dohmen et al. (2010), which reports that Li diffuses 10^{5-7} times faster than Fe-Mg in olivine at 800° C.

Low-Ca pyroxenes are the most Li-enriched phases identified in our study (Figs. 5b,d,f). The LPx of group 1 chondrules (Fig. 5b) demonstrates a significant range of [Li] and δ^7 Li between two components (0.4 μ g·g⁻¹, +33‰ for component 1 and 3 μ g·g⁻¹, -33‰ for component 2). LPx^{irim} forms a cluster with an average [Li] of 7 μ g·g⁻¹ and δ^7 Li of -11‰. The existence of the array of LPx and the cluster of LPx^{irim} is also observed on group 2 (Fig. 5d).

The partition coefficients for Li between olivine and the melt and between low-Ca pyroxenes and melt are estimated to be from 0.01 to 1.04, and 0.13 to 0.22, respectively, at 0.1–3000 MPa and 1320–1390°C (Tomascak et al., 2016). Using these values, the partition coefficient for Li (k_{Li}) between olivine and low-Ca pyroxene is estimated as being 0.1 to 5. From the observations of this study we find the [Li] of Oli^{rim} and LPx^{irim} to be 0.2 and 7 μ g · g⁻¹, respectively. The enrichment of Li in LPx^{irim} goes against element partitioning under an equilibrium condition during asteroidal processing. The high [Li] and low δ^7 Li observed in LPx^{irim} from both group 1 and group 2 ([Li] and $\delta^7 \text{Li} = 7 \,\mu\text{g} \cdot \text{g}^{-1}$ and -11%, respectively) should reflect those of the melt during igneous-rim formation. The Li-isotope composition of the final melt on igneous-rim formation ($\delta^7 \text{Li} \sim -11\%$) is coincidentally consistent with that of the initial chondrule melt (δ' Li $\sim -10\%$) estimated from group 1 olivines. Evaporation of Li from chondrule melt and recondensation of Li to the final melt could explain the reprise of the isotope composition. However, the igneous rim is characterized by high abundances of not only alkalines, but also Si relative to the main region of the chondrules (Rubin and Wasson, 1987). Krot et al. (2004) suggested that igneous rims formed by either gas-solid condensation of silica-normative materials onto chondrule surfaces and subsequent incomplete melting, or by direct condensation of gas, enriched in alkalines and SiO. Thus the enrichment of Li in LPx^{irim} resulted from a gaseous reservoir that may not be genetically related with the chondrule melt. The Li-isotope composition of the gaseous reservoir is estimated to be $\delta^7 \text{Li} \sim -11\%$. During chondrule formation the $\delta^7 \text{Li}$ of LPx was approaching the Li-isotope composition of the gaseous reservoir, indicating that the chondrule melt interacted with the gaseous reservoir during chondrule formation.

4.2.3 Li-isotope distribution within chondrule groups

Ol^{irim} of group 1 is depleted in Li (Fig. 6a) and surrounded by LPx^{irim} enriched in Li. Li-isotope compositions were equilibrated among Ol^{irim} but Li concentrations between Ol^{irim} and LPx^{irim} were out of equilibration, as mentioned previously. These observations, together with O-isotope composition and the anhedral shape of Ol^{irim}, suggest that Li was diffusing out from Ol^{irim}, as the size of Ol^{irim} was becoming smaller. The duration, of igneous-rim formation for group 1, was sufficiently long to allow equilibration of Li isotopes, but not O isotopes.

For group 2, it is not obvious if Li isotopes of Ol were reaching equilibrium during igneous-rim formation (Fig. 6b), because of the lack of analyses in Ol^{irim}. Further analyses are required.

The Li of LPx^{irim} in group 3 chondrules differs to that in group 1 and 2 (Fig. 5f). The concentration of LPx^{irim} Li is low, but its isotope composition is consistent, when compared to whole rock values. Perhaps the Li concentration and isotope composition of the gas during igneous-rim formation differs from those of group 1 and group 2. Ol^{irim} tends to have a higher Fe/Mg ratio relative to that of Ol, thus the Fe/Mg ratios of olivine could have been modified during igneous-rim formation. However, both Ol and Ol^{irim} maintain Li-isotope variation. We conclude that unlike the igneous-rim formation process of group 1 chondrules, the igneous-rim formation process of group 3 did not equilibrate Li isotopes. We do not find pure galactic cosmic ray components with $^7\text{Li}/^6\text{Li}\sim 2$ that corresponds to $\delta^7\text{Li}\sim -800\%$ (Read and Viola, 1984; Ramaty et al., 1996; Meneguzzi et al., 1971) in group 3 olivines. Therefore, group 3 olivines do not maintain the Li-isotope signature of the interstellar medium, from before the formation of the solar nebula, although they do maintain the O-isotope variation inherited from precursors.

4.3. Li isotope compositions of chondrule precursors

Considering that chondrules experienced multiple heating events (e.g., Jones et al., 2005) and asteroidal alteration, the ⁷Li/⁶Li heterogeneity could be caused by high temperature fractional crystallization, devolatilization, and diffusion (Tomascak et al., 2016). Although Chaussidon and Robert (1998) assumed the isotope heterogeneities observed in ordinary chondrite chondrules were inherited only from precursors, our combined O-

and Li-isotope study of the Allende chondrules can distinguish between the Li-isotope heterogeneity produced before, during and after chondrule formation.

Group 3 chondrules contain relict olivines which are overgrown by olivine, which crystallized from chondrule melts and their range of O-isotope compositions is inherited from precursors. Also group 3 olivines maintain variations in Li isotopes of the precursors that formed prior to chondrule formation and igneous-rim formation. Since the number of analyses for chondrule PO-3n is limited (Fig. 4i), we focus on group 3 chondrule PO-8 that preserves significant variation of both O- and Li-isotope compositions. The δ^7 Li as a function of Δ^{17} O for group 3 chondrule PO-8 is shown in Figure 7. Olivines in group 3 chondrule PO-8 have different Δ^{17} O and δ^{7} Li values, which reflects either distinct relicts or melt. We see that variation of δ^7 Li differs depending on values of Δ^{17} O. The Li-isotope composition varies from δ^{7} Li -23% to -3% with an O-isotope composition close to that of refractory inclusions ($-25 < \Delta^{17}O < -20\%$) and is relatively constant ($-8 \pm 4\%$), when the O-isotope composition is in between $\Delta^{17}O$ -20%and -5%. Note that average Li-isotope compositions of group 3 olivines ($\delta^7 \text{Li} \sim -8\%$) is consistent with that of the chondrule melt ($\delta^7 \text{Li} \sim -10\%$) estimated from group 1 olivines. We infer that even with a constant $\Delta^{17}O$ (-25 < $\Delta^{17}O$ < -20\%), $\delta^{7}Li$ is distributed heterogeneously, probably reflecting kinetic processes.

We evaluate if it is possible that the heterogeneities of Δ^{17} O and δ^7 Li resulted from measurements of 16 O-rich relict-olivine and 16 O-poor olivine with overgrowths, which the probe overlapped. Depending on the relative fraction of the two domains, the array of δ^{18} O/ δ^{17} O on a three O-isotope diagram could be produced. If this is the case, the Li-isotope composition of 16 O-rich relict olivine is estimated to be δ^7 Li -23%. Chaussidon et al. (2006) analyzed refractory inclusions that do not show evidence for post-melting redistribution of Li and estimated δ^7 Li to be $-24 \pm 46\%$. The average of δ^7 Li values of refractory inclusions is consistent with the relict olivine end component. If the variation of Δ^{17} O is dominated by a relative fraction of relict olivine and olivine with overgrowths on probing, we would expect to see mixing between δ^7 Li of the end components (-23%) and δ^7 Li of the chondrule melt (-10%; estimated from group 1 olivines). However, we see no monotonic correlation in Δ^{17} O- δ^7 Li space (Fig. 7). We suggest that the variations of Δ^{17} O and δ^7 Li are not dominated by probing and they reflect distinct relicts or melt having different O- and Li-isotope compositions either changing with time or having local heterogeneity.

Chaussidon and Robert (1998) reported Li-isotope variation preserved in ordinary chondrite chondrules ($-12.7 < \delta^7 \text{Li} < 34.5\%$) and proposed that the variation is inherited from chondrule precursors that preserved signatures resulting from the mixing of two different nucleosynthetic sources (galactic cosmic rays and big-bang nucleosynthesis). The variation is consistent with the range observed in group 1 olivines (from $\delta^7 \text{Li} - 10 \text{ to } +40\%$) and could result from the chondrule formation process. The lack of $\delta^7 \text{Li}$ values below -10% in ordinary chondrite chondrules can be explained because or-

dinary chondrite chondrules do not have the ¹⁶O-rich signature of refractory inclusions (Kita et al., 2010).

We conclude that the $\delta^7 \text{Li}$ values of olivine, which formed prior to the asteroid, reflect a combination of: (1) variation of $\delta^7 \text{Li}$ from -23 to -3% inherited from precursors with O-isotope composition of refractory inclusions ($-25 < \Delta^{17} \text{O} < -20\%$), (2) relatively constant $\delta^7 \text{Li} - 8 \pm 4\%$ inherited from precursors with O isotopes in the rage $-20 < \Delta^{17} \text{O} < -5\%$, (3) homogenization of $\delta^7 \text{Li}$ in the melt during chondrule formation with $\delta^7 \text{Li} \sim -10\%$, (4) modification of $\delta^7 \text{Li}$ in the melt by devolatilization up to $\delta^7 \text{Li} \sim +40\%$, and (5) homogenization of $\delta^7 \text{Li}$ during igneous-rim formation.

4.4. Lithium and oxygen reservoirs in the solar nebula

It has been suggested that gas-melt interactions played an important role during the formation of magnesium-rich chondrules. Libourel et al. (2006) inferred that typical type I chondrules are objects composed of an inherited igneous component (mainly forsteritic olivine with varying amounts of Fe,Ni-metal) and a second igneous component (low-Ca pyroxene, high-Ca pyroxene, and glass) formed through interaction with the nebula gas. Tissandier et al. (2002) demonstrated experimentally that partially molten chondrule-like samples exposed to gaseous SiO incorporate silica, inducing the crystallization of low-Ca pyroxene. These experiments suggest that various features observed in many type I chondrules, such as the presence of low-Ca pyroxene in the igneous rim of chondrules or the partial resorption of olivine, can be obtained by varying the duration or the temperature of the gas-melt interaction. High partial pressures of SiO (g) in the solar nebula have been invoked to explain the apparent enrichment of low-Ca pyroxene relative to olivine at the periphery of porphyritic chondrules.

Chaussidon et al. (2008) analyzed olivines and pyroxenes of CR and CV chondrules and estimated the O-isotope composition of the nebular gas to be $\Delta^{17}O$ –0.1‰. It was inferred that the O-isotope compositions indicate that olivines in type I chondrules are relict grains that formed prior to the last chondrule formation. Chaussidon et al. (2008) also state that interaction between the gas and melt (forming pyroxenes) during chondrule formation was in an open system. Harju et al. (2014) observed a sub-‰ difference in δ^{29} Si between olivine and low-Ca pyroxene in CV and CR chondrules and concluded that the difference reflects Si isotopic condensation in the nebula under near-equilibrium conditions. Their model proposes that all olivines are relict, being either inherited from an earlier generation of igneous bodies, or from an earlier generation of chondrules, and that pyroxenes are produced by interaction of chondrule melt with the gas. In contrast, there are many chondrules in which it is clear that both olivine and pyroxene grew from the same melt during chondrule cooling, and Rudraswami et al. (2011), Ushikubo et al. (2012), and Tenner et al. (2013, 2015) have shown that there is no difference in O-isotope compositions between olivine and pyroxene in chondrules.

Our O- and Li-isotope observations suggest that olivines and low-Ca pyroxenes

formed from the same melt. Subsequently, low-Ca pyroxenes formed from the melt while interacting with a gaseous reservoir. The finding is inconsistent with the existence of a gaseous reservoir with a constant isotope composition during the period of chondrule formation. Rubin (2017) argued that the petrologic observation made on Semarkona is controversial for later stage gas-melt interaction and insisted that the formation of low-Ca pyroxene in the periphery region does not require gas-melt interaction. We predict that Li analyses of low-Ca pyroxenes in the periphery region will reveal the existence of gas-melt interaction during formation of ordinary chondrite chondrules.

We summarize Li reservoirs in the solar nebula inferred from this study (Fig. 8). Prior to chondrule formation, olivines with a different O-isotope composition were formed at different times or in different places. There were isotope reservoirs with δ^7 Li that were varying from -23 to -3\% and with the Δ^{17} O of refractory inclusions. During chondrule formation, the precursors were melted and O- and Li-isotope compositions were homogenized. Due to the high solidus temperature, olivine solidified first from the melt. Before low-Ca pyroxene crystallized, the O-isotope composition of the melt could have changed by a few \%. Li-isotope composition of the melt changed from $\delta' \text{Li} \sim -10\%$ to +40% by devolatilization. A portion of olivines and low-Ca pyroxene subsequently crystallized from isotopically heavy melt ($\delta^7 \text{Li} \sim +40\%$). At the end of chondrule formation (during crystallization of low-Ca pyroxene), the Li-isotope composition of the melt changed drastically via interaction with a gaseous reservoir that was enriched in alkalines and Si with $\delta^7 \text{Li} \sim -11\%$. Then on igneous-rim formation, low-Ca pyroxenes formed from melt that was enriched in Li. A mechanical mixture of objects with differing Li results in an asteroidal reservoir corresponding to whole rock Allende δ' Li $\sim 2.4\%$. After accumulation into the asteroid, plagioclase and a portion of olivine obtained O- and Li-isotope compositions reflecting the asteroidal reservoir.

We are now aware of existence of a distinct reservoir, as suggested by Seitz et al. (2007), with $\delta^7 \text{Li} - 11\%$ in the early Solar system. Lithium-isotope compositions of chondrite, achondrite, lunar, and Martian meteorites obtained by whole-rock analyses range from $\delta^7 \text{Li} - 2$ to 6% (Tomascak et al., 2016) and none of these can serve as the reservoir. It has been suggested that a primordial water reservoir existed in the early solar system (Sakamoto et al., 2007). Whilst the oxygen isotopic signatures identified by the current study are different from those of this theoretical reservoir, a similarly water rich reservoir could be the source of the Li-isotope composition predicted here. It is not clear if the reservoir still exists in the current Solar system.

5. CONCLUSIONS

We carried out 258 pairs of in situ O- and Li-isotope analyses for olivine, low-Ca pyroxene, high-Ca pyroxene, and plagioclase from the main bodies of type IAllende (CV3) chondrules and their igneous rims. We suggest that the isotope compositions

of plagioclases and secondary ferroan olivine (Fo₆₂–Fo₆₅)resulted from asteroidal processes. Their Li-isotope compositions are similar to that of whole-rock Allende. After excluding these olivines and plagioclases, by comparison of O, Li, and Fe/Mg, we suggest that the Li-isotope composition of most chondrules was modified during igneous-rim formation, but not significantly by asteroidal process.

By analyzing a chondrule, which lacks evidence of equilibration in both O- and Li-isotope compositions during the period of chondrule and igneous-rim formation, we demonstrate a correlation between $\Delta^{17}O$ and $\delta^{7}Li$ preserved in olivine. We found that $\delta^{7}Li$ varied from -23 to -3‰ for a given $\Delta^{17}O$ of refractory inclusions. Based on the Li-isotope composition of low-Ca pyroxene formed during chondrule formation and igneous-rim formation, we suggest that chondrule melt reacted with a gaseous reservoir with $\delta^{7}Li \sim -11\%$.

Li concentrations and isotope compositions of chondrules from the Allende chondrite resulted from the multiple layers of processing that the chondrules have undergone. The variation of Li-isotope compositions reflects (1) variation of $\delta^7 \text{Li}$ from -23 to -3% inherited from precursors with the O-isotope composition of refractory inclusions ($-25 < \Delta^{17} \text{O} < -20\%$), (2) relatively constant $\delta^7 \text{Li} - 8 \pm 4\%$ inherited from precursors with O isotopes that range by $-20 < \Delta^{17} \text{O} < -5\%$, (3) homogenization of $\delta^7 \text{Li}$ in the melt during chondrule formation with $\delta^7 \text{Li} \sim -10\%$, (4) modification of $\delta^7 \text{Li}$ in the melt by devolatilization up to $\delta^7 \text{Li} \sim +40\%$, and (5) homogenization of $\delta^7 \text{Li}$ during igneous-rim formation.

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Table 1: Major-element compositions of chondrule constituents.

		phase	occurrence	n	compo*	$[SiO_2]$	[TiO]	[Al ₂ O ₃]	$[Cr_2O_3]$	[FeO]	[MnO]	[MgO]	[CaO]	[Na ₂ O]	$[K_2O]$	[NiO]	$[SO_3]$	$[P_2O_5]$	[F]	[Cl]	total
group 1	BOP-2	Ol _{>Fo95}	inner BO	7	F099	42.86	0.08	0.19	0.13	1.48	0.13	54.91	0.19	_	_	0.02	0.02	0.01	0.01	0.01	100.03
		$Ol_{>Fo95}$	shell of inner BO	34	Fo ₉₈	42.62	0.08	0.26	0.24	2.00	0.10	54.55	0.21	0.01	_	0.02	0.01	0.02	0.01	_	100.12
		$Ol_{< Fo95}$	shell of inner BO	7	Fo ₉₃	41.69	0.07	0.26	0.24	6.87	0.10	50.59	0.16	0.02	-	0.02	0.01	0.01	_	_	100.03
		Ol _{>Fo95}	inner BO	2	Fo ₉₈	42.70	0.07	0.23	0.16	2.42	0.08	54.27	0.18	0.01	_	_	0.01	0.02	_	_	100.15
		$Ol_{>Fo95}$	shell of outer BOP	17	Fo ₉₇	42.60	0.08	0.30	0.25	2.82	0.10	53.79	0.20	0.03	_	0.02	0.01	0.01	0.01	_	100.24
		$Ol_{$	shell of outer BOP	1	Fo ₉₅	42.13	0.05	0.26	0.20	5.28	0.06	51.99	0.17	_	_	0.03	0.02	_	_	_	100.20
		Ol ^{irim}	igneous rim	2	Fo ₉₁	41.29	0.03	0.17	0.08	8.78	0.04	49.21	0.15	0.01	_	0.01	0.01	-	0.01	-	99.77
		LPx	outer BOP	41	$En_{97}Wo_2$	59.14	0.26	1.63	0.50	0.41	0.07	37.40	1.11	0.01	0.01	0.02	0.02	0.02	0.01	_	100.61
		LPx ^{irim}	igneous rim	7	$En_{98}Wo_1$	59.70	0.13	0.86	0.48	0.67	0.11	38.11	0.55	-	_	0.01	0.02	0.01	0.01	-	100.67
		HPx	outer BOP	6	$En_{68}Wo_{31}$	55.58	0.96	3.41	0.72	0.38	0.12	24.05	15.44	0.01	_	0.01	0.02	-	0.02	-	100.72
		Pl	outer BOP	10	$An_{82}Ab_{18}$	48.70	0.05	30.86	0.04	0.45	0.03	1.58	16.45	2.00	-	0.02	0.01	_	0.03	-	100.21
	PO-6	Ol _{>Fo95}	main	2	F099	42.99	0.09	0.35	0.24	1.35	0.02	54.51	0.41	-	_	-	-	-	_	-	99.97
		$Ol_{$	main	4	Fo ₈₉	40.98	0.05	0.26	0.20	10.62	0.15	47.38	0.22	0.01	0.01	0.06	0.02	0.06	_	_	100.00
		Ol ^{irim}	igneous rim	3	Fo ₈₄	40.42	0.02	0.14	0.06	14.45	0.18	44.19	0.16	-	_	0.07	0.01	0.03	_	-	99.74
		Ol ^{FeO}	main	2	Fo ₆₄	37.11	0.01	1.26	0.40	29.61	0.19	29.19	0.16	0.09	0.01	0.05	0.09	0.08	0.01	-	98.26
		LPxirim	igneous rim	12	$En_{97}Wo_1$	59.73	0.14	0.99	0.55	1.11	0.08	37.96	0.50	0.01	-	0.03	0.02	0.02	0.02	-	101.15
	BO-4	Ol _{>Fo95}	main	10	Fo ₉₈	43.10	0.05	0.23	0.24	1.98	0.06	54.43	0.26	0.02	_	0.03	_	0.02	0.01	_	100.44
		Ol _{<fo95< sub=""></fo95<>}	shell	3	Fo ₉₂	41.95	0.04	0.20	0.26	7.50	0.10	49.73	0.13	0.01	0.01	0.03	0.01	0.02	0.01	-	99.99
group 2	PO-7	Ol _{>Fo95}	main	2	Fo ₉₈	42.97	0.04	0.24	0.18	1.88	0.04	54.39	0.23	0.02	-	0.04	0.01	0.06	_	_	100.08
		$Ol_{< Fo95}$	main	1	Fo ₉₀	41.33	_	0.14	0.07	9.94	0.05	47.65	0.20	_	0.01	_	_	_	0.04	_	99.41
		LPx ^{irim}	igneous rim	3	$En_{98}Wo_1$	59.58	0.21	1.49	0.60	0.76	0.11	37.49	0.58	-	0.01	-	0.03	0.04	0.01	-	100.90
	PO-1	Ol _{>Fo95}	main	4	F099	42.96	0.04	0.29	0.11	1.36	0.01	54.63	0.49	_	_	0.02	0.01	0.07	0.01	_	100.01
		$Ol_{< Fo95}$	main	1	Fo ₉₃	41.06	0.07	0.23	0.16	6.61	0.08	50.31	0.45	0.01	_	0.07	0.01	_	0.03	_	99.07
		Ol ^{FeO}	main	1	Fo ₆₅	30.13	0.62	13.06	0.25	26.42	0.15	27.41	0.70	0.15	-	0.15	0.26	0.06	_	0.02	99.39
		LPx	main	3	$En_{93}Wo_5$	58.45	0.26	1.29	0.66	1.48	0.12	36.10	2.49	_	_	0.02	0.01	_	0.01	_	100.88
		HPx	main	3	$En_{56}Wo_{43}$	50.71	1.31	9.38	1.11	0.61	0.22	18.22	19.10	0.02	_	0.02	0.01	_	_	_	100.70
	PO-3c	Ol _{<fo95< sub=""></fo95<>}	main	3	Fo ₈₅	40.33	0.04	0.22	0.16	14.05	0.10	45.01	0.23	0.01	_	0.03	0.01	0.04	_	_	100.23
		Ol ^{FeO}	main	1	Fo ₆₂	36.66	0.10	0.18	0.43	32.92	0.22	30.43	0.19	_	_	_	0.01	0.06	_	_	101.32
	PO-3s	Ol _{>Fo95}	main	2	Fo ₉₈	43.04	0.03	0.26	0.20	2.02	0.04	54.70	0.45	_	_	0.05	0.01	_	_	0.01	100.76
		Ol _{<fo95< sub=""></fo95<>}	main	3	Fo ₉₁	41.67	0.03	0.24	0.18	8.25	0.04	49.36	0.30	-	0.01	0.01	-	0.02	0.02	-	100.11
group 3	PO-8	Ol _{<fo95< sub=""></fo95<>}	core	3	Fo ₈₇	40.85	0.02	0.29	0.28	12.26	0.10	45.57	0.38	0.01	-	0.12	0.03	_	_	_	99.90
		Ol _{>Fo95}	mantle	12	Fo ₉₆	42.52	0.02	0.17	0.12	3.73	0.11	52.80	0.26	0.01	_	0.13	0.02	0.03	0.01	_	99.93
		$Ol_{< Fo95}$	mantle	22	Fo ₉₂	41.93	0.02	0.19	0.09	7.47	0.10	49.90	0.27	0.01	_	0.04	0.02	0.03	0.01	_	100.07
		Ol ^{irim}	igneous rim	10	Fo ₈₇	41.09	0.02	0.22	0.05	12.21	0.11	45.89	0.27	-	_	0.05	0.01	0.03	0.01	_	99.96
		LPxirim	igneous rim	2	$En_{93}Wo_6$	59.71	0.23	1.26	0.67	0.99	0.13	34.65	3.00	-	_	0.06	0.04	0.03	0.03	_	100.78
		HPx ^{irim}	igneous rim	4	$En_{62}Wo_{37}$	54.82	0.79	4.00	0.83	0.72	0.13	21.72	17.92	0.01	_	0.03	0.02	0.01	0.02	_	101.02
		Pl	mantle	3	$An_{82}Ab_{18}$	48.33	0.04	30.78	0.04	1.29	0.01	0.79	16.63	2.01	0.01	0.03	0.01	-	_	_	99.97
	PO-3n	Ol _{>Fo95}	main	1	Fo ₉₅	42.62	0.01	0.14	0.01	4.44	0.16	52.41	0.17	_	_	0.05	_	-	0.05	_	100.04
		Ol _{<fo95< sub=""></fo95<>}	main	4	Fo ₈₉	41.45	0.02	0.14	0.05	10.28	0.10	47.50	0.19	_	_	0.04	0.01	0.01	0.02	0.01	99.77

Element concentration is expressed in wt%. Element concentration not available is denoted by —. Abbreviation compo* denotes major-element composition and is defined by $\frac{F_0}{100} \equiv \frac{Mg}{Mg+Fe}$, $\frac{En|W_0}{100} \equiv \frac{Mg|Ca}{Mg+Fe+Ca}$, and $\frac{Ab|An}{100} \equiv \frac{Na|Ca}{Ca+Na+K}$ in molar units. Ol, LPx, and HPx are olivine, low-Ca pyroxene, and high-Ca pyroxene at the main chondrule (including mantle or core regions). Ol_{>Fo95} and Ol_{<Fo95} are Ol with chemical composition Fo_{>95} and Fo₆₅₋₉₅.

Ol^{irim}, LPx^{irim}, and HPx^{irim} are olivine, low-Ca pyroxene, and high-Ca pyroxene at igneous rims.

Pl is plagioclase and Ol^{FeO} is olivine with chemical composition Fo_{≤65}.

Table 2: O- and Li-isotope compositions of chondrule constituents.

		phase	occurrence	n	compo*	$\delta^{18}{ m O}$	$\delta^{17}{ m O}$	$\Delta^{17}O$	δ^7 Li	[Li] ^{μg·g¯}
group 1	BOP-2	Ol _{>Fo95}	inner BO	7	Fo _{99.0(4)}	-5 (1)	-7.7 (7)	-5.4 (4)	22 (6)	0.5(2)
		$Ol_{>Fo95}$	shell of inner BO	34	Fo ₉₈₍₁₎	-5.0 (8)	-7.9 (6)	-5.3 (5)	6 (4)	0.7(4)
		$Ol_{< Fo95}$	shell of inner BO	7	Fo ₉₃₍₃₎	-4.8 (6)	-7.9 (5)	-5.4(3)	7 (4)	0.8(4)
		$Ol_{>Fo95}$	outer BOP	2	Fo ₉₈₍₁₎	-4.6 (2)	-7.9 (4)	-5.5 (3)	17 (14)	0.3(1)
		$Ol_{>Fo95}$	shell of outer BOP	17	Fo ₉₇₍₁₎	-4.6 (9)	-7.7 (5)	-5.3 (4)	9 (4)	0.2(2)
		$Ol_{< Fo95}$	shell of outer BOP	1	Fo ₉₅	-6.0	-8.4	-5.2	-5	0.2
		Ol ^{irim}	igneous rim	2	Fo ₉₁	-4.8 (1)	-7.9 (3)	-5.4(2)	5 (1)	0.2(1)
		LPx	outer BOP	41	$En_{97(2)}Wo_{2(2)}$	-3.1 (7)	-7.0 (6)	-5.4 (4)	-4 (13)	3(2)
		LPxirim	igneous rim	7	$En_{98.0(3)}Wo_{1.0(2)}$	-3.1 (7)	-7.0 (6)	-5.4 (4)	-4 (5)	5.2(4)
		HPx	outer BOP	6	En ₆₈₍₆₎ Wo ₃₁₍₆₎	-2(1)	-6.3 (9)	-5.1 (5)	-6 (6)	0.6(3)
		Pl	outer BOP	10	$An_{82(1)}Ab_{18(1)}$	-2.5 (9)	-6.9 (9)	-5.6 (5)	1 (4)	0.0(0)
	PO-6	Ol _{>Fo95}	main	2	Fo ₉₉₍₁₎	-6(1)	-9 (2)	-6(1)	-3 (7)	0.7(3)
		$Ol_{< Fo95}$	main	4	Fo ₈₉₍₃₎	-4.2 (6)	-7.1 (3)	-5.0(3)	5 (3)	0.2(1)
		Ol ^{irim}	igneous rim	3	Fo ₈₄₍₆₎	-4.4(3)	-7.0 (4)	-4.7(3)	6 (6)	0.1(1)
		Ol^{FeO}	main	2	Fo ₆₄₍₁₎	4.8(2)	0(2)	-3(1)	2(2)	0.2(2)
		LPxirim	igneous rim	12	En ₉₇₍₁₎ Wo _{0,9(2)}	-3.7 (6)	-7.0 (6)	-5.0(3)	-15 (6)	8 (3)
	BO-4	Ol>Fo95	main	10	Fo ₉₈₍₁₎	-4.4 (6)	-7.8 (7)	-5.5 (5)	11 (12)	0.1(1)
		Ol _{<fo95< sub=""></fo95<>}	shell	3	Fo ₉₂₍₂₎	-4.8 (1)	-7.6 (3)	-5.1 (3)	9 (9)	0.3(2)
group 2	PO-7	Ol _{>Fo95}	main	2	Fo _{98.0(1)}	-5.9 (4)	-9.2 (6)	-6.1 (4)	-7 (11)	1.1 (6)
		$Ol_{< Fo95}$	main	1	Fo ₉₀	-6.3	-9.3	-6.0	10	0.2
		LPxirim	igneous rim	3	En _{98.0(2)} Wo _{1.1(2)}	-5.7 (2)	-9.1 (5)	-6.2(4)	-13 (3)	7.7 (9)
	PO-1	Ol _{>Fo95}	main	4	Fo ₉₉₍₁₎	-6.2 (7)	-9.9(2)	-6.7(3)	4 (12)	0.2(2)
		$Ol_{$	main	1	Fo ₉₃	-6.9	-9.3	-5.7	4	0.4
		OlFeO	main	1	Fo ₆₅	5.8	0.84	-2.2	-2	0.1
		LPx	main	3	En ₉₃₍₂₎ Wo ₅₍₁₎	-5.2 (3)	-9.1 (6)	-6.4(5)	4 (13)	3(1)
		HPx	main	3	En ₅₆₍₂₎ Wo ₄₃₍₂₎	-6.4 (6)	-9.7(2)	-6.4(2)	5 (9)	0.4(2)
	PO-3c	$Ol_{< Fo95}$	main	3	Fo ₈₅₍₇₎	-6.8 (8)	-9.8 (4)	-6.3 (4)	-6 (6)	0.1(0)
		Ol ^{FeO}	main	1	Fo ₆₂	-2.7	-4.5	-3.1	5	0.0
	PO-3s	Ol _{>Fo95}	main	2	Fo ₉₈₍₂₎	-6.6 (3)	-9.7 (3)	-6.3(2)	-11 (10)	0.4(1)
		Ol _{<fo95< sub=""></fo95<>}	main	3	Fo ₉₁₍₄₎	-6.7 (5)	-9.2 (5)	-5.7 (4)	2 (7)	0.5(2)
group 3	PO-8	Ol _{<fo95< sub=""></fo95<>}	core	3	Fo ₈₇₍₅₎	-17 (6)	-20 (6)	-11 (3)	-7 (1)	0.2(1)
8 - 1		$Ol_{>Fo95}$	mantle	12	Fo ₉₆₍₁₎	-28 (15)	-30 (14)	-15 (6)	-6 (5)	0.2(1)
		Ol _{<fo95< sub=""></fo95<>}	mantle	22	Fo ₉₂₍₂₎	-34 (14)	-35 (13)	-18 (6)	-9 (4)	0.2(2)
		Ol ^{irim}	igneous rim	10	Fo ₈₇₍₄₎	-27 (18)	-29 (17)	-15 (8)	-12 (7)	0.3(2)
		LPxirim	igneous rim	2	$En_{93(1)}Wo_{6(1)}$	-2(2)	-6(1)	-5.5 (1)	-1 (3)	0.9(5)
		HPx ^{irim}	igneous rim	4	$En_{62(4)}Wo_{37(4)}$	-5.4 (4)	-8.1 (6)	-5.3 (6)	-9 (7)	0.2(1)
		Pl	mantle	3	$An_{82(1)}Ab_{18(1)}$	3 (2)	-2 (1)	-3.3 (4)	-4 (6)	0.1 (0)
	PO-3n	Ol _{>Fo95}	main	1	Fo ₉₅	-19	-21	-11	36	0.2
		Ol _{<fo95< sub=""></fo95<>}	main	4	Fo ₈₉₍₈₎	-18 (8)	-20 (8)	-11 (3)	5 (1)	0.1 (1)

Abbreviation compo* denotes major-element composition. Variation among n analyses (1SD) is shown in parenthesis.

Ol, LPx, and HPx are olivine, low-Ca pyroxene, and high-Ca pyroxene at the main chondrule (including mantle or core regions).

 $Ol_{>Fo95}$ and $Ol_{<Fo95}$ are Ol with chemical composition $Fo_{>95}$ and Fo_{65-95} .

Ol^{irim}, LPx^{irim}, and HPx^{irim} are olivine, low-Ca pyroxene, and high-Ca pyroxene at igneous rims.

Pl is plagioclase and Ol^{FeO} is olivine with chemical composition $Fo_{\leq 65}$.

Table 3: Summary of O- and Li-isotope compositions of chondrule constituents.

	phase	n	compo*	$\delta^{18}{ m O}$	$\delta^{17}\mathrm{O}$	Δ^{17} O	δ^7 Li	$[\mathrm{Li}]^{\mu\mathrm{g}\cdot\mathrm{g}^{-1}}$
group 1	Ol _{>Fo95}	72	Fo ₉₈₍₁₎	-4.8 (8)	-7.8 (6)	-5.4 (5)	9 (8)	0.5 (4)
	$Ol_{< Fo95}$	15	Fo ₈₇₍₅₎	-4.7 (7)	-7.7 (5)	-5.2 (3)	6 (6)	0.5(4)
	Ol ^{irim}	5	$Fo_{91(2)}$	-4.5 (3)	-7.3 (6)	-5.0 (4)	5 (4)	0.2(1)
	LPx	41	$En_{97(4)}Wo_{2(4)}$	-3.1 (7)	-7.0 (6)	-5.4 (4)	-4 (13)	3 (2)
	LPx^{irim}	19	$En_{98(1)}Wo_{1(0)}$	-3 (1)	-6.7 (8)	-5.1 (4)	-11 (8)	7 (3)
	HPx	6	$En_{69(8)}Wo_{30(8)}$	-2.5 (9)	-6.9 (9)	-5.6 (5)	-6 (6)	0.6(3)
	Pl	10	$An_{81(2)}Ab_{19(2)}$	5 (2)	-1 (1)	-3.3 (5)	1 (4)	0.04(4)
	Ol^{FeO}	2	Fo ₆₄₍₁₎	4.8 (2)	0(2)	-3 (1)	2(2)	0.2(2)
group 2	Ol _{>Fo95}	8	Fo ₉₈₍₁₎	-6.2 (6)	-9.7 (4)	-6.4 (4)	-3 (12)	0.5 (5)
	$Ol_{< Fo95}$	8	Fo ₈₉₍₆₎	-6.7 (5)	-9.5 (5)	-6.0 (4)	0(7)	0.3(2)
	LPx	3	$En_{94(1)}Wo_{5(1)}$	-5.2 (3)	-9.1 (6)	-6.4 (5)	4 (13)	3 (1)
	LPxirim	3	$En_{99(1)}Wo_{1(1)}$	-5.7 (2)	-9.1 (5)	-6.2 (4)	-13 (3)	7.7(9)
	HPx	3	$En_{53(4)}Wo_{46(4)}$	-6.4 (6)	-9.7 (2)	-6.4 (2)	5 (9)	0.4(2)
	$\mathrm{Ol}^{\mathrm{FeO}}$	2	Fo ₆₃₍₃₎	2 (6)	-2 (4)	-2.6 (6)	1 (5)	0.08(4)
group 3	Ol _{>Fo95}	13	Fo ₉₆₍₁₎	-27 (14)	-29 (13)	-15 (6)	-3 (12)	0.20(7)
	$Ol_{< Fo95}$	29	Fo ₉₁₍₄₎	-30 (14)	-31 (14)	-16 (6)	-7 (6)	0.2(2)
	Ol^{irim}	10	Fo ₈₇₍₄₎	-27 (18)	-29 (17)	-15 (8)	-12 (7)	0.3(2)
	LPx^{irim}	2	$En_{93(0)}Wo_{6(1)}$	-2 (2)	-6(1)	-5.5 (1)	-1 (3)	0.9(5)
	HPx^{irim}	4	$En_{64(5)}Wo_{34(6)}$	-5.4 (4)	-8.1 (6)	-5.3 (6)	-9 (7)	0.20(6)
	Pl	3	$An_{83(2)}Ab_{17(2)}$	3 (2)	-2(1)	-3.3 (4)	-4 (6)	0.08 (4)

Abbreviation compo* denotes major-element composition. Variation within group (1SD) is shown in parenthesis.

Ol, LPx, and HPx are olivine, low-Ca pyroxene, and high-Ca pyroxene at the main chondrule (including mantle or core regions). $Ol_{>F095}$ and $Ol_{<F095}$ are Ol with chemical composition $Fo_{>95}$ and Fo_{65-95} .

Ol^{irim}, LPx^{irim}, and HPx^{irim} are olivine, low-Ca pyroxene, and high-Ca pyroxene at igneous rims.

Pl is plagioclase and Ol^{FeO} is olivine with chemical composition Fo_{≤65}.

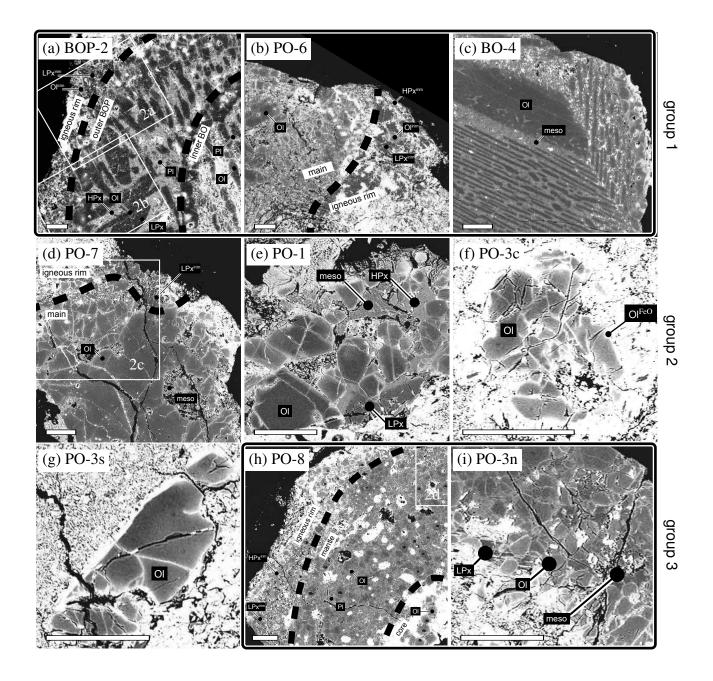


Figure 1: Backscattered electron images of (a, b, c) group 1 chondrules BOP-2, PO-6, and BO-4, (d, e, f, g) group 2 chondrules PO-7, PO-1, PO-3c, and PO-3s, and (h, i) group 3 chondrules PO-8 and PO-3n. Width of scale bar is $100 \, \mu \text{m}$.

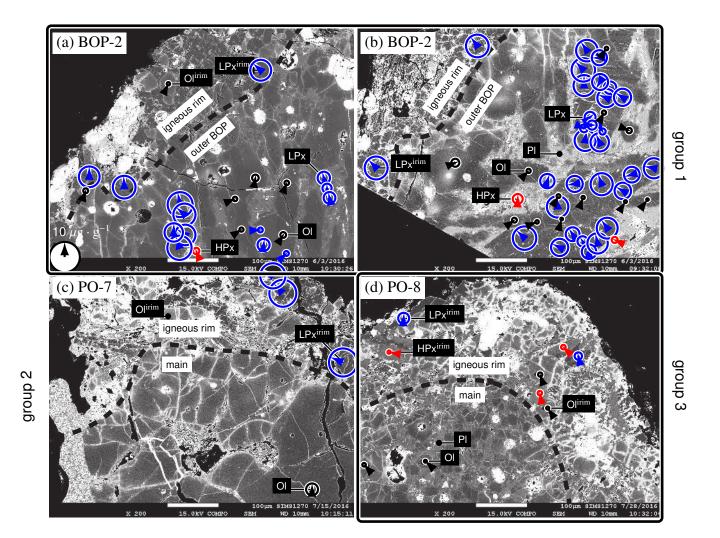


Figure 2: Li element-and-isotope compositions superimposed on BSE images: (a,b) group 1 chondrule BOP-2, (c) group 2 chondrule PO-7, and (d) group 3 chondrule PO-8. Element concentration is proportional to the area of the circle and isotope composition is shown by angle of needle. Angle of nine, twelve, and three o'clock correspondences to $\delta^7 \text{Li}$ –20, 0, and +20%, respectively. A reference circle shown on left-and-bottom corner of (a) corresponds to [Li] $10 \ \mu\text{g} \cdot \text{g}^{-1}$ and $\delta^7 \text{Li}$ 0%.

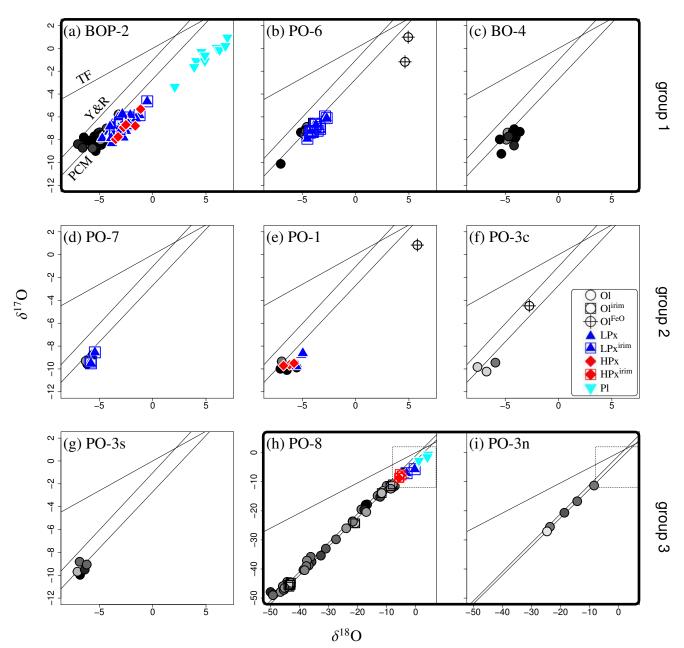


Figure 3: O-isotope compositions of constituents of (a, b, c) group 1 chondrules BOP-2, PO-6, and BO-4, (d, e, f, g) group 2 chondrules PO-7, PO-1, PO-3c, and PO-3s, and (h, i) group 3 chondrules PO-8 and PO-3n. Olivines are shown by circle. Olivines with chemical composition $Fo_{\leq 65}$ (Ol^{FeO}) are shown with a cross symbol. The Fe/Mg ratio is indicated by the color of the circle. White and black circles correspond to Fo_{70} and Fo_{100} , respectively. Low-Ca pyroxenes, high-Ca pyroxenes, and plagioclase are shown by triangle (blue), diamond (red), and inversed triangle (cyan), respectively. Olivines, low-Ca pyroxenes, and high-Ca pyroxenes in igneous rims (Ol^{irim}, LPx^{irim}, and HPx^{irim}) are shown with symbol of square.

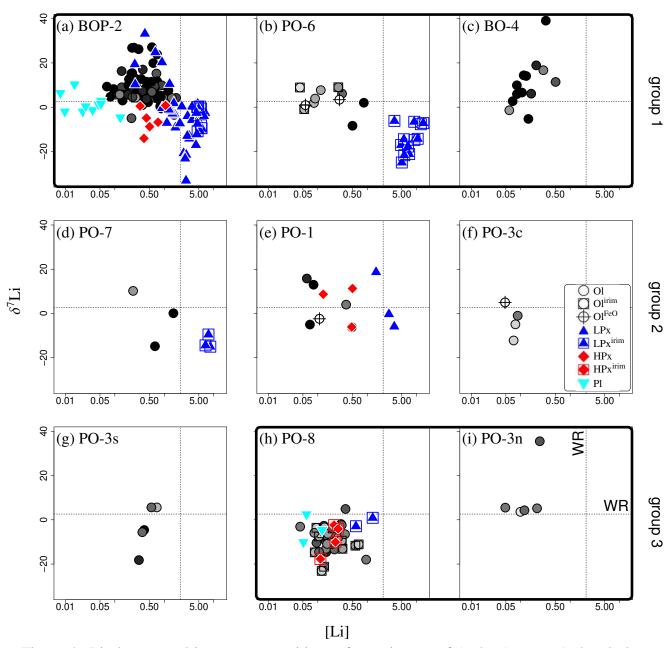


Figure 4: Li element-and-isotope compositions of constituents of (a, b, c) group 1 chondrules BOP-2, PO-6, and BO-4, (d, e, f, g) group 2 chondrules PO-7, PO-1, PO-3c, and PO-3s, and (h, i) group 3 chondrules PO-8 and PO-3n.

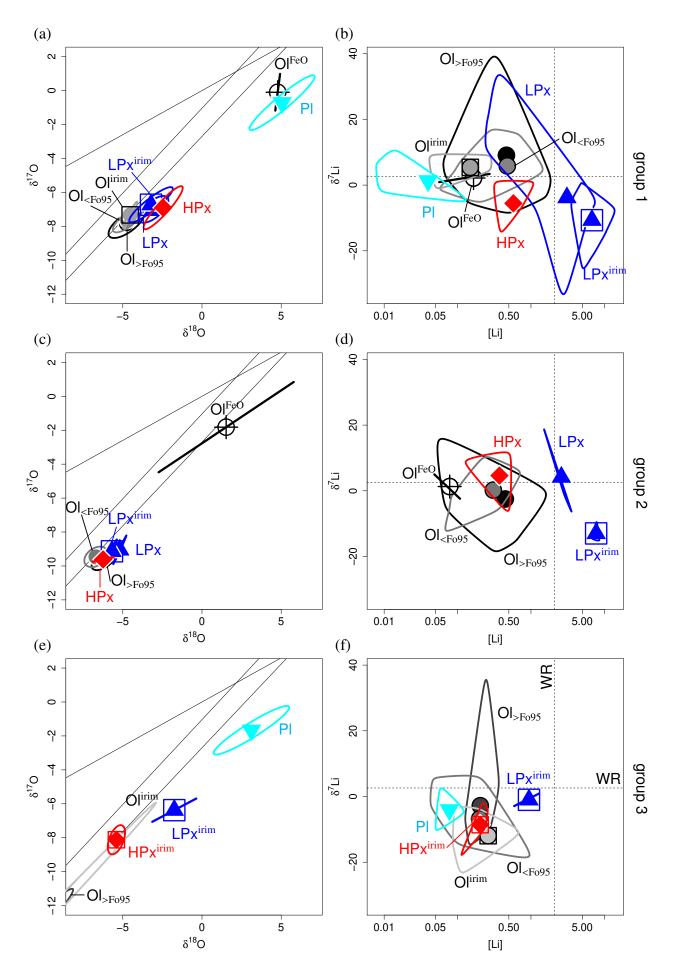


Figure 5: O-isotope and Li-element and -isotope compositions of constituents of (a, b) group 1 chondrule, (c, d) group 2 chondrule, and (e, f) group 3 chondrule. For each constituent and region an average of datasets is shown.

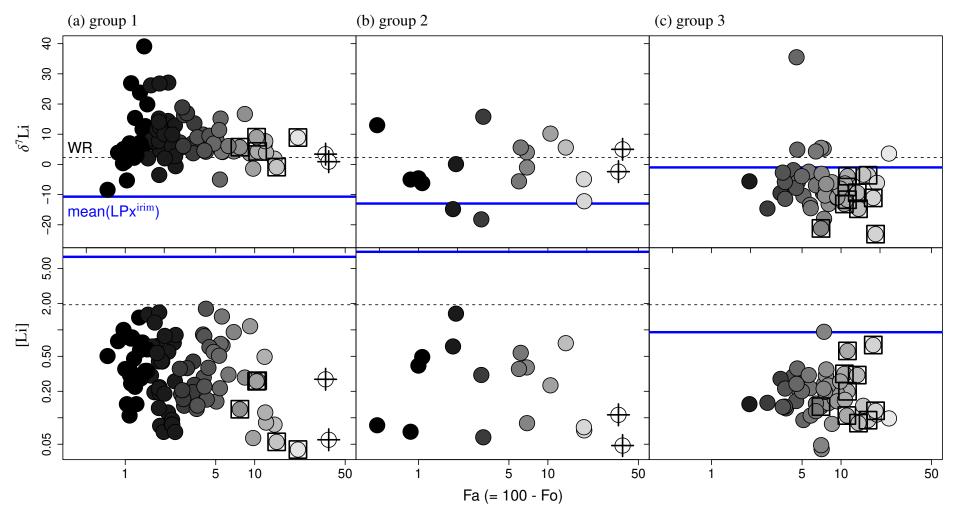


Figure 6: Li element-and-isotope compositions of olivines as a function of the Fe/Mg ratio. Olivines of (a) chondrule group 1, (b) chondrule group 2, and (c) chondrule group 3 are shown. Olivine that occurred in igneous rims (Ol^{irim}) and those with chemical composition $Fo_{\leq 65}$ (Ol^{FeO}) are shown with symbols of square and cross, respectively.

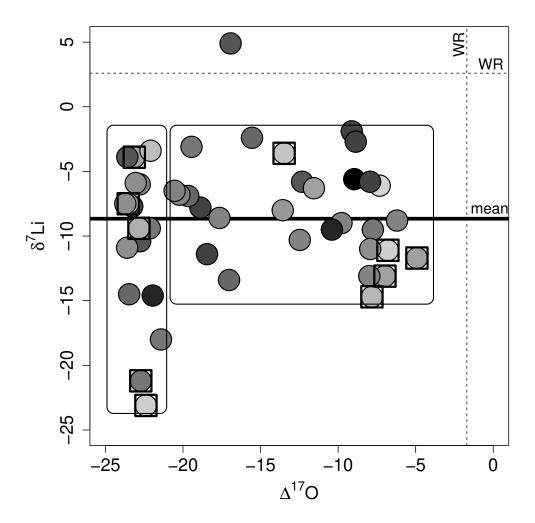


Figure 7: O- and Li-isotope compositions of group 3 chondrule PO-8: The Fe/Mg ratio is shown by color of circle. WR and mean indicate whole-rock Allende and the mean of olivine from group 3 chondrule PO-8, respectively.

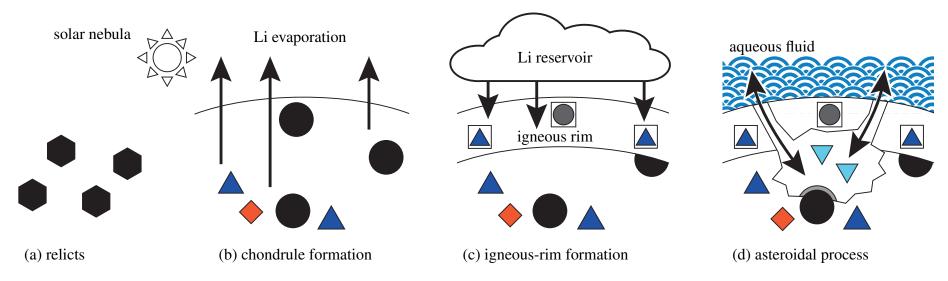


Figure 8: Schematic illustration of Li-isotope evolution.

Appendix 1 RESULTS

All datasets presented in this manuscript are included on our institutional depository DREAM, which is open to the public.

Appendix 1.1. Matrix effect correction on isotope analyses

Matrix effects on instrumental mass fractionation have been known to occur for oxygen-and lithium-isotope measurements by using SIMS. Therefore, we estimated the matrix effects of olivine, low-Ca pyroxene, high-Ca pyroxene, and plagioclase, relative to working standards, based on the isotope measurements of reference olivines, pyroxenes, plagioclases, and synthetic glasses with certain extents of compositional varieties in terms of $X_{\rm Mg}$ ($\equiv \frac{[{\rm Mg}]^{\rm mol}}{[{\rm Fe}]^{\rm mol}+[{\rm Mg}]^{\rm mol}+[{\rm Kg}]^{\rm mol}+[{\rm Kg}]^{$

Elemental and isotope compositions of the reference materials analyzed in this study are summarized in Table S1, where major element concentrations were determined by EPMA (JEOL JXA-8800 and JXA-8530F) and FE-SEM (JEOL JSM-7001F) equipped with EDS and Oxford AZtec X-Max and X-act. O-isotope compositions were determined by CO₂ laser fluorination and by using a Finnigan MAT253 mass spectrometer (Tanaka and Nakamura, 2013). Li concentrations and isotope compositions were determined by the method described in Moriguti and Nakamura (1998), except that Li-isotope measurements were carried out using a multicollector-inductively coupled plasma-mass spectrometer, Thermo-Finnigan Neptune (Tang et al., 2007).

Matrix effects on oxygen-isotope analysis Matrix effects on O-isotope analysis of olivine (Fo₆₂₋₁₀₀) relative to the working standard ol-sc (Fo₉₁) were examined by analyses of reference olivines (Fo₄₈₋₁₀₀). The estimated matrix effects of olivine (Fo₇₀₋₁₀₀) were within spot-to-spot reproducibility on the working standard ol-sc (δ^{18} O 0.2%; 1SD). Thus we did not apply the matrix effect correction for olivines with Fo₇₀₋₁₀₀. Whereas for olivines with Fo₆₂₋₇₀, the matrix effects relative to ol-sc were significant (Table S2 and Fig. S1a). Thus we corrected the matrix effect (δ^{18} O from -0.6 to 0%) on olivines with Fo₆₂₋₇₀.

Matrix effects of low-Ca pyroxene ($En_{90-98}Wo_{1-9}$) and high-Ca pyroxene ($En_{54-74}Wo_{26-45}$) relative to the working standard ol-sc were examined by analyses of reference low-Ca pyroxenes ($Wo_{0.3-2}$) and high-Ca pyroxenes (Wo_{37-50}), and chemical-composition dependence was confirmed (Table S2 and Fig. S1b). We corrected the matrix effects on low-Ca pyroxene ($\delta^{18}O$ from -2.2 to -1.7%0) and high-Ca pyroxene ($\delta^{18}O$ from -0.6 to +0.7%0).

Matrix effect of plagioclase $(Ab_{17-19}An_{81-83})$ relative to the working standard olsc was estimated by analyses of reference plagioclases (Ab_{40-98}) and synthetic glasses (Ab_{53-100}) , to be $\delta^{18}O$ from +0.1 to +0.2‰, from the chemical-composition dependence (Table S2 and Fig. S1c). Since the matrix effects of plagioclases $(Ab_{17-19}An_{81-83})$ were comparable to spot-to-spot reproducibility on ol-sc $(\delta^{18}O \sim 0.2\%; 1SD)$, we did not apply the matrix effect correction to plagioclase $(Ab_{17-19}An_{81-83})$.

For any phases examined in this study, the matrix effects were estimated consistently with compositional variations of the phases. By the correction, the matrix-related systematic deviations in analyses were significantly reduced ($\leq 0.4\%$). Although uncertainty on the matrix effect correction is controlled also by spot-to-spot reproducibility on each reference material, the standard deviations (1SD) on reference olivines, pyroxenes, plagioclases, and glasses range from 0.1 to 0.4%. Accordingly we estimated the total

uncertainty including that on the matrix effect correction to be $\pm 0.4\%$, which is comparable to or slightly larger than the internal standard error $(1\sigma_m)$ on individual analysis $(\pm 0.1 \sim 0.3\%)$ in δ^{18} O; Table S4).

Matrix effects on lithium element and isotope analysis Li concentrations were calculated using secondary ion-intensity relative to primary beam-current ($^7\text{Li}^+/\text{I}_{PRI}$), and a correction factor k ($\frac{\mu g \cdot g^{-1}}{\text{cps/nA}}$) estimated by analyses of working standards, ol-sc1 and en-sl1. The correction factor could change from session to session thus the working standards were analyzed for each session.

Representative secondary ion intensities of ${}^7\mathrm{Li}^+$ were $4{-}9{\times}10^4$ cps for ol-sc1 ([Li] ${}^{\mu\mathrm{g}\cdot\mathrm{g}^{-1}}{=}1.26$) and en-sl1 ([Li] ${}^{\mu\mathrm{g}\cdot\mathrm{g}^{-1}}{=}29.2$) at primary beam currents of 1–15 nA, and typical 1SD errors on estimation of elemental concentrations were 8–13%. The correction factor changed depending on the primary beam-current, and the apparent elemental concentrations of the working standards were deviated to some extent from the reference values of the working standards; typically $\leq 25\%$, but up to $\sim 50\%$ for the low-[Li] standard by the low primary beam-current.

Matrix effects on Li-isotope analysis of olivine relative to ol-sc1 (Fo₉₁), of low-Ca pyroxene and high-Ca pyroxene relative to en-sl1 (En₉₈Wo_{0.1}), and of plagioclase (Ab₁₅₋₃₇An₆₃₋₈₅) relative to ol-sc1 were estimated by analyses of reference olivines, pyroxenes, and plagioclases, respectively. As for olivine, we estimated the matrix effects, and corrected them for olivine (Fo₆₂₋₁₀₀) from +2.6 to -0.7% relative to ol-sc1 (Table S3 and Figure S2a). Matrix effects of low-Ca pyroxene (En₉₀₋₉₈Wo₁₋₉) and high-Ca pyroxene (En₅₄₋₇₄Wo₂₆₋₄₅) were estimated to be from +0.0 to +0.4‰, and from +1.0 to +1.8‰, respectively (Table S3). However, spot-to-spot reproducibility on the working standard en-sl1 was 2.6‰ in 1SD (Fig. S2b), thus we did not apply the matrix effect correction. As for plagioclase, matrix effects were estimated to be from -0.6 to -0.6‰ (Table S3), but spot-to-spot reproducibility on the working standard ol-sc1 (0.6‰ in 1SD) was comparable to the estimated matrix effects (Fig. S2c), thus we did not apply the matrix effect correction.

For any phases examined in this study, the matrix effects were estimated consistently with compositional variations of the phases, and the matrix-related systematic bias in Li-isotope analysis was successfully corrected. Thus uncertainty on the matrix effect correction is controlled mainly by the spot-to-spot reproducibility on reference materials. The standard deviations on reference olivines, pyroxenes, and plagioclases range from 0.6 to 2.6% (1SD), thus we estimated the total uncertainty on the matrix effect correction to be $\pm 3\%$, which is comparable to the internal standard error $(1\sigma_m)$ on individual analysis (typically $\pm 1 \sim 3\%$ and occasionally up to 6% in $\delta^7 \text{Li}$; Table S4).

Appendix 1.2. Oxygen-isotope compositions

O-isotope compositions are shown in Table S4. O-isotope compositions of each chondrule are detailed in this section.

BOP-2, PO-6, and BO-4: Average Δ^{17} O values of olivines (Fo_{>65} in mantle) of the chondrules are $-5.4 \pm 0.6\%$ (± 1 SD). Average Δ^{17} O values for MgO-rich olivines (Fo_{>95} in main region) of BOP-2, PO-6, and BO-4 are -5.5, -6.0, and -5.5%, respectively.

BOP-2 consists of inner BO, outer BOP, and an igneous rim, as inferred already. We refer to the bar-dominated area and shell-dominated area of the BO as inner BO and shell of inner BO, respectively. We also refer to bar-dominated areas and shell-dominated areas of the BOP as outer BOP and shell of outer BOP, respectively. The BOP-2 is composed of five layers (inner BO, shell of inner BO, outer BOP, shell of outer BOP, and igneous rim) and contains four silicate phases (olivine, high-Ca pyroxene, low-Ca pyroxene, and plagioclase). δ^{18} O and Δ^{17} O values for all olivines (Fo_{>65}) range from -7.0 to -3.2% and from -6.2 to -4.1%, respectively. Average δ^{18} O values for olivines (Fo_{>65}) in inner BO, shell of inner BO, outer BOP, shell of outer BOP, and igneous rim are -4.6, -4.9, -4.6, -4.7, and -4.8%, respectively. Average Δ^{17} O values for olivines (Fo_{>65}) in inner BO, shell of inner BO, outer BOP, shell of outer BOP, and igneous rim are -5.4, -5.3, -5.5, -5.3, and -5.4%, respectively. High-Ca pyroxenes occur only in outer BOP. δ^{18} O and Δ^{17} O values for high-Ca pyroxenes range from -3.5 to -1.1% and from -6.2 to -4.7%, respectively. Low-Ca pyroxenes occur in outer BOP and igneous rims. δ^{18} O values for low-Ca pyroxenes in outer BOP and igneous rims range from -4.7 to -1.6 and from -3.5 to $-0.5\%_0$, respectively. Δ^{17} O values for low-Ca pyroxenes in outer BOP and igneous rims range from -6.4 to -4.3 and from -5.7to -4.4\%, respectively. O-isotope compositions of high-Ca and low-Ca pyroxenes are heavier than those of olivines δ^{18} O and Δ^{17} O values for plagioclase range from +2.1 to +7.0% and from -4.5 to -2.7%, respectively.

PO-6 consists of two layers (main and igneous rims). The main region is dominated by olivines and igneous rim is dominated by olivines and low-Ca pyroxenes. Average δ^{18} O and Δ^{17} O values for olivines (Fo_{>65}) in main region range from -7.0 to -3.6% and from -6.5 to -4.5%, respectively. Average δ^{18} O and Δ^{17} O values for olivines (Fo_{>65}) in igneous rim range from -4.6 to -4.0% and from -4.9 to -4.4%, respectively. δ^{18} O and Δ^{17} O values for an olivine with Fo₉₉ in main region are -7.0 and -6.5%, respectively. Average of (δ^{18} O, Δ^{17} O) for two FeO-rich olivines (Fo₆₃ and Fo₆₅) is (+4.8%, -2.6%). Low-Ca pyroxenes occur only in igneous rim. δ^{18} O and Δ^{17} O values for low-Ca pyroxene range from -4.5 to -2.7% and from -5.6 to -4.5%, respectively. Except for three olivines (one olivine Fo₉₉ and two FeO-rich olivines), O-isotope compositions of olivine and low-Ca pyroxene are consistent.

BO-4 consists of two layers (main and shell regions). The main region consists of barred olivines and mesostasis, and the shell region is dominated by olivine. δ^{18} O values

for olivines in the main region (Fo_{>95}) and olivines in the shell region (Fo_{>65}) range from -5.5 to -3.6% and from -4.9 to -4.8%, respectively. O-isotope composition of olivines in the shell region is similar to those in the main region.

PO-7, PO-1, PO-3c, and PO-3s: The O-isotope compositions of olivines (Fo_{>65}) of the chondrules are $\Delta^{17}O$ –6.2 ± 0.4% (±1SD). Average $\delta^{18}O$ values for olivines (Fo_{>65}) of PO-7, PO-1, PO-3c, and PO-3s, are –6.0, –6.3, –6.8, and –6.7‰, respectively. Average $\Delta^{17}O$ values for the olivines (Fo_{>65}) of PO-7, PO-1, PO-3c, and PO-3s, are –6.1, –6.5, –6.3, and –6.0‰, respectively. Average $\delta^{18}O$ and $\Delta^{17}O$ values for all analyses made on the olivines in the main regions of the four chondrules (Fo_{>65}) are –6.5 and –6.2‰, respectively.

PO-7 consists of main region dominated by olivines and igneous rim dominated by low-Ca pyroxenes. $\delta^{18}O$ and $\Delta^{17}O$ values for olivines (Fo_{>65}) range from -6.3 to -5.6‰ and from -6.4 to -5.9‰, respectively. $\delta^{18}O$ and $\Delta^{17}O$ values for low-Ca pyroxenes range from -5.8 to -5.4‰ and from -6.5 to -5.7‰, respectively. O-isotope composition of low-Ca pyroxene on the igneous rim is consistent with those on main region.

PO-1 consists of olivine (Fo_{>65}), FeO-rich olivine (Fo_{≤65}), high-Ca pyroxene, and low-Ca pyroxene. $\delta^{18}O$ and $\Delta^{17}O$ values for olivines (Fo_{>65}) range from -7.0 to -5.5% and from -7.0 to -5.7%, respectively. $\delta^{18}O$ and $\Delta^{17}O$ values for an FeO-rich olivine is +5.8 and -2.2%, respectively. $\delta^{18}O$ and $\Delta^{17}O$ values for high-Ca pyroxenes range from -6.8 to -5.8% and from -6.5 to -6.2%, respectively. $\delta^{18}O$ and $\Delta^{17}O$ values for low-Ca pyroxenes range from -5.5 to -5.0% and from -6.9 to -6.0%, respectively.

PO-3c consists of olivine (Fo_{>65}) and FeO-rich olivine (Fo₆₂). δ^{18} O and Δ^{17} O values for olivines (Fo_{>65}) range from -7.6 to -5.9% and from -6.7 to -5.9%, respectively. δ^{18} O and Δ^{17} O values for an FeO-rich olivine are -2.7 and -3.1%, respectively.

PO-3s consists of olivine (Fo_{>65}). δ^{18} O and Δ^{17} values for olivines range from -7.0 to -6.2% and from -6.4 to -5.3%, respectively.

PO-8 and PO-3n: The O-isotope compositions of olivine in two chondrules (PO-8 and PO-3n) vary significantly, and δ^{18} O and Δ^{17} O values range from -49.9 to -3.8% and from -23.7 to -4.9%, respectively.

PO-8 consists of three layers (core, mantle, and igneous rims) with four silicates (olivine, high-Ca pyroxene, low-Ca pyroxene, and plagioclase). δ^{18} O values for olivines (Fo₈₁₋₉₉) in core, mantle, and igneous rims range from -23.9 to -11.3‰, from -49.9 to -7.2‰, and from -43.8 to -3.8‰, respectively. Δ^{17} O values for the olivines in core, mantle, and igneous rim range from -13.6 to -7.3‰, from -23.7 to -6.2‰, and from -23.5 to -4.9‰, respectively. Variation for O-isotope composition of olivines in core region (Fo₈₁₋₉₀) is small relative to olivine in mantle and igneous rims (Fo₈₂₋₉₈). High-Ca and low-Ca pyroxenes only occur in igneous rims. δ^{18} O values for high-Ca and

low-Ca pyroxenes range from -5.7 to -4.8% and from -3.1 to -0.4%, respectively. $\Delta^{17}\mathrm{O}$ values for high-Ca and low-Ca pyroxenes range from -5.7 to -4.5% and from -5.5 to -5.4%, respectively. O-isotope compositions of high-Ca and low-Ca pyroxenes are more $^{16}\mathrm{O}$ -depleted than that of olivines. $\delta^{18}\mathrm{O}$ and $\Delta^{17}\mathrm{O}$ values for plagioclase range from +1.1 to +4.3% and from -3.6 to -2.9%, respectively. O-isotope composition of plagioclases is the heaviest among silicates.

PO-3n is dominated by olivine (Fo_{77–95}). δ^{18} O and Δ^{17} O values for the olivines range from -24.6 to -8.4% and from -14.3 to -6.9%, respectively.

Appendix 1.3. Lithium concentrations and isotope compositions

Li concentration and isotope compositions are shown in Table S4. Li concentration and isotope compositions of each chondrule are detailed in this section.

BOP-2 consists of five layers (inner BO, shell of inner BO, outer BOP, shell of outer BOP, and igneous rim) with four silicates (olivine, low-Ca pyroxene, high-Ca pyroxene, and plagioclase), as inferred already. [Li] and δ^7 Li of all olivines range from 0.069 to 1.74 μ g · g⁻¹ and from -5.0 to +27.1%, respectively. [Li] of olivines in inner BO, shell of inner BO, outer BOP, shell of outer BOP, and igneous rim range from 0.22 to $0.72 \ \mu g \cdot g^{-1}$, from 0.14 to 1.74 $\mu g \cdot g^{-1}$, from 0.20 to 0.30 $\mu g \cdot g^{-1}$, from 0.069 to 0.64 $\mu g \cdot g^{-1}$, and from 0.12 to 0.26 $\mu g \cdot g^{-1}$, respectively. $\delta^7 \text{Li}$ values for olivines in inner BO, shell of inner BO, outer BOP, shell of outer BOP, and igneous rims range from +11.7 to +27.1%, from -3.5 to +17.0%, from +7.0 to +26.2%, from -5.0 to +16.3%, and from +4.3 to +5.7\%, respectively. [Li] and δ^7 Li values for an olivine (Fo₉₈) are $0.56 \,\mu\mathrm{g}\cdot\mathrm{g}^{-1}$ and +27.1%. [Li] and $\delta^7\mathrm{Li}$ values for high-Ca pyroxene occurring in outer BOP range from 0.32 to 1.1 μ g · g⁻¹ and from -14.1 to +0.8\%, respectively. Low-Ca pyroxenes are subdivided into two types based on occurrences in either outer BOP or igneous rim. [Li] values for low-Ca pyroxenes in outer BOP and igneous rim range from 0.25 to 6.7 μ g · g⁻¹ and 4.6 to 5.7 μ g · g⁻¹, respectively. δ ⁷Li values for low-Ca pyroxenes in outer BOP and igneous rim range from -33.2 to +33.2% and from -10.9to +0.3\%, respectively. [Li] and δ^7 Li values of plagioclase range from 0.0079 to 0.13 $\mu g \cdot g^{-1}$ and from -4.6 to +10.3%, respectively. The plagioclase is the most Li-depleted mineral in BOP-2. Its element abundance is one order of magnitude smaller than that of bulk samples and δ^7 Li values for the plagioclase are similar to those for the bulk samples (1.93 μ g · g⁻¹ and +2.4‰; Seitz et al., 2012).

PO-6 consists of two layers (main and igneous rim) dominated by three minerals [olivine (Fo_{>65}), FeO-rich olivine (Fo_{≤65}), and low-Ca pyroxene], as inferred already. [Li] values for olivines (Fo_{>65}) in main and igneous rim range from 0.084 to 0.86 μ g·g⁻¹ and from 0.044 to 0.26 μ g·g⁻¹, respectively. δ^7 Li values for olivines (Fo_{>65}) in main and igneous rim range from –8.4 to +7.7‰ and from –0.9 to +9.0‰, respectively. [Li] and δ^7 Li values for an olivine (Fo₉₉) are 0.50 μ g·g⁻¹ and –8.4‰, respectively. The Liisotope composition of the olivine is the lightest among olivines (Fo_{>65}). Average [Li]

and $\delta^7 \text{Li}$ values for FeO-rich olivines (Fo₆₃₋₆₅) are 0.17 $\mu g \cdot g^{-1}$ and +2.1‰, respectively. Li-isotope composition of the FeO-rich olivine is similar to that of bulk. [Li] and $\delta^7 \text{Li}$ values for low-Ca pyroxenes in igneous rim range from 3.5 to 13.6 $\mu g \cdot g^{-1}$ and from -25.0 to -6.2‰, respectively.

BO-4 consists of two layers (main and shell) dominated by olivine (Fo_{>65}), as inferred already. [Li] values for olivines (Fo_{>65}) in main and shell regions range from 0.069 to 0.32 μ g · g⁻¹ and from 0.059 to 0.51 μ g · g⁻¹, respectively. δ^7 Li values for olivines (Fo_{>65}) in main and shell regions range from –5.3 to +39.1‰ and from –1.4 to +16.7‰, respectively. [Li] and δ^7 Li values for an olivine (Fo₉₉) are 0.32 μ g · g⁻¹ and 39.1‰. The Li-isotope composition of the olivine is the heaviest.

PO-7 consists of two layers (main and igneous rim) dominated two minerals [olivine (Fo_{>65}) and low-Ca pyroxene], as inferred already. [Li] and δ^7 Li values for olivines (Fo₉₀₋₉₈) range from 0.23 to 1.53 μ g · g⁻¹ and from -14.8 to +10.2‰. [Li] and δ^7 Li values for low-Ca pyroxenes in igneous rim range from 6.8 to 8.5 μ g · g⁻¹ and from -15.0 to -9.5‰, respectively. δ^7 Li value for an olivine (Fo₉₈) is -14.8‰, whereas average δ^7 Li value for other two olivines (Fo₉₀₋₉₉) is +5 ± 5‰.

PO-1 consists of four silicates [olivine (Fo_{>65}), FeO-rich olivine (Fo_{≤65}), high-Ca pyroxene, and low-Ca pyroxene], as inferred already. [Li] and δ^7 Li values for olivines (Fo_{>65}) range from 0.06 to 0.49 μ g · g⁻¹ and from -6.2 to +15.8‰, respectively. [Li] values for high-Ca and low-Ca pyroxenes range from 0.13 to 0.50 μ g · g⁻¹ and from 1.5 to 3.5 μ g · g⁻¹, respectively. δ^7 Li values for high-Ca and low-Ca pyroxenes range from -6.1 to +11.3‰ and from -5.9 to +18.7‰, respectively. [Li] and δ^7 Li values for an FeO-rich olivine (Fo₆₅) are 0.11 μ g · g⁻¹ and -2.4‰, respectively. Average [Li] values for minerals increase in order of FeO-rich olivine (Fo₆₅, 0.11 μ g · g⁻¹), olivines (Fo_{>65}, 0.2 μ g · g⁻¹), high-Ca pyroxene (0.4 μ g · g⁻¹), and low-Ca pyroxene (2.6 μ g · g⁻¹).

PO-3c is dominated by olivines, as inferred already. A portion of olivines is classified as FeO-rich (Fo₆₂) olivines. [Li] and δ^7 Li values for olivines (Fo₈₁₋₉₃) range from 0.072 to 0.087 μ g · g⁻¹ and from -12.2 to -1.0‰, respectively. [Li] and δ^7 Li values for the FeO-rich olivine (Fo₆₂) are 0.048 μ g · g⁻¹ and +5.0‰, respectively.

PO-3s is dominated by olivines (Fo_{>65}), as inferred already. [Li] and δ^7 Li values for the olivines range from 0.31 to 0.71 μ g · g⁻¹ and from -18.2 to +5.6‰, respectively. δ^7 Li value for an olivine (Fo₉₇) is significantly light (-18.2‰).

PO-8 consists of three layers and four silicates, as inferred already. [Li] values for olivines (Fo_{>65}) in core, mantle, and igneous rim range from 0.11 to 0.26 μ g · g⁻¹, from 0.044 to 0.95 μ g · g⁻¹, and from 0.087 to 0.67 μ g · g⁻¹, respectively. δ^7 Li values for the olivines in core, mantle, and igneous rim range from -8.0 to -6.1‰, from -18.0 to +4.9‰, and from -23.1 to -3.6‰, respectively. Average [Li] values for olivines in three layers are consistent but average δ^7 Li values decrease from core to igneous rim. [Li] values for high-Ca and low-Ca pyroxenes range from 0.11 to 0.26 μ g · g⁻¹ and from 0.59 to 1.3 μ g · g⁻¹, respectively. δ^7 Li values for high-Ca and low-Ca pyroxenes range

from -17.7 to -2.4% and from -2.9 to +0.9%, respectively. [Li] and $\delta^7 \text{Li}$ values for plagioclase range from 0.051 to $0.12~\mu\text{g}\cdot\text{g}^{-1}$ and from -10.1 to +2.5%, respectively. Average [Li] values for minerals increase in order of plagioclase $(0.08~\mu\text{g}\cdot\text{g}^{-1})$, olivines $(0.2~\mu\text{g}\cdot\text{g}^{-1})$ in three occurrences), high-Ca pyroxene $(0.2~\mu\text{g}\cdot\text{g}^{-1})$, and low-Ca pyroxene $(0.9~\mu\text{g}\cdot\text{g}^{-1})$.

PO-3n is dominated by olivines (Fo_{>65}), as inferred already. [Li] and δ^7 Li values for olivines range from 0.049 to 0.24 μ g · g⁻¹ and from +3.6 to +35.5‰. δ^7 Li value of an olivine (Fo₉₅) is +35.5‰, whereas average δ^7 Li value for other four olivines (Fo_{77–94}) is +4.7‰.

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Table S1: Elemental and isotope compositions of reference materials.

				-10 -	-17 -	-7- •	
reference material	$X_{ m Mg}$	X_{Ca}	X_{Na}	δ^{18} O	δ^{17} O	δ^7 Li	$[\mathrm{Li}]^{\mu\mathrm{g}\cdot\mathrm{g}^{-1}}$
olivine							
ol-sc	0.91	_	_	5.22	2.68	_	_
ol-AY	1.00	_	_	2.85	1.45	_	-
ol-fo	0.99	_	_	23.6	12.4	_	_
ol-LS1-A	0.70	_	_	5.34	2.80	_	_
ol-NWA6704(b)	0.48	_	_	4.09	1.12	_	_
ol-sc1	0.91	_	_	_	_	2.93	1.26
ol-fo1	0.99	_	_	_	_	5.57	29.0
ol-fa1	0.01	_	_	_	_	14.8	28.8
low-Ca pyroxene							
opx-tz1001	0.91	0.003	_	10.8	5.62	_	_
opx-klb1	0.90	0.02	_	5.73	3.01	_	_
en-sl1	0.98	0.001	_	_	_	1.86	29.2
high-Ca pyroxene							
cpx-sax33	0.72	0.37	_	5.85	3.03	_	_
cpx-klb1	0.90	0.46	_	5.42	2.82	_	_
cpx-di-tz1001	1.00	0.50	_	20.6	10.7	_	_
cpx-sae113	0.84	0.31	_	_	_	0.14	3.76
cpx-sae116	0.84	0.34	_	_	_	-7.52	9.02
cpx-sae152	0.82	0.39	_	_	_	1.74	4.85
cpx-sax39c	0.82	0.33	_	_	_	3.47	3.05
plagioclase							
pl-alb21245	_	_	0.98	10.6	5.56	_	_
pl-lab	_	_	0.40	6.43	3.38	_	_
pl-anorthoclase-c	_	_	0.72	_	_	2.2	21.6
pl-bytownite-c2s	_	_	0.42	_	_	-38.5	8.04
synthetic glass							
gl-ab100	_	_	1.00	18.5	9.39	_	_
gl-an25ab75	_	_	0.78	17.4	8.91	_	_
gl-an50ab50	_	_	0.53	16.5	8.45	_	_
							·

Major element compositions, shown by X_{Mg} ($\equiv \frac{[\mathrm{Mg}]^{\mathrm{mol}}}{[\mathrm{Fe}]^{\mathrm{mol}} + [\mathrm{Mg}]^{\mathrm{mol}}}$), X_{Ca} ($\equiv \frac{[\mathrm{Ca}]^{\mathrm{mol}}}{[\mathrm{Fe}]^{\mathrm{mol}} + [\mathrm{Mg}]^{\mathrm{mol}}}$), and X_{Na} ($\equiv \frac{[\mathrm{Na}]^{\mathrm{mol}}}{[\mathrm{Ca}]^{\mathrm{mol}} + [\mathrm{Na}]^{\mathrm{mol}} + [\mathrm{Kg}]^{\mathrm{mol}}}$), were determined by EPMA and SEM-EDS. O- and Liisotope compositions were determined by laser fluorination and gas mass spectrometry, and by wet chemistry with ICP-MS, respectively. See text for analytical details.

Table S2: Matrix effects relative to a working standard for O isotope analysis.

phase	formula	$^{ m calc}\Delta\delta^{18}{ m O}_{ m ws}^{ m phase}$	remark
olivine (Fo _{70–100})	$\Delta \delta^{18} O_{\text{ol-sc}}^{\text{phase}} \equiv 0$	≡ 0	no correction
olivine (Fo ₆₂₋₇₀)	$\Delta \delta^{18} O_{\text{ol-sc}}^{\text{phase}} = 6.9 \times (X_{\text{Mg}} - 0.7)$	-0.6 to $+0.0%$	correction applied
low-Ca pyroxene $(En_{90-98}Wo_{1-9})$	$\Delta \delta^{18} O_{\text{ol-sc}}^{\text{phase}} = 6.7 \times X_{\text{Ca}} - 2.3$	-2.2 to $-1.7%$	correction applied
high-Ca pyroxene (En ₅₄₋₇₄ Wo ₂₆₋₄₅)	$\Delta \delta^{18} O_{\text{ol-sc}}^{\text{phase}} = 6.7 \times X_{\text{Ca}} - 2.3$	-0.6 to $+0.7%$	correction applied
plagioclase $(Ab_{17-19}An_{81-83})$	$\Delta \delta^{18} O_{\text{ol-sc}}^{\text{phase}} = -5.8 \times X_{\text{Na}} + 1.2$	+0.1 to +0.2\%	no correction

Table S3: Matrix effects relative to working standards for Li isotope analysis.

phase	formula	$^{\rm calc}\Delta\delta^7{\rm Li}_{\rm ws}^{\rm phase}$	remark
olivine (Fo _{62–100})	$\Delta \delta^7 \text{Li}_{\text{ol-sc1}}^{\text{phase}} = -9.1 \times (X_{\text{Mg}} - 0.91)$	+2.6 to -0.8\%	correction applied
low-Ca pyroxene (En _{90–98} Wo _{1–9})	$\Delta \delta^7 \text{Li}_{\text{en-sl1}}^{\text{phase}} = 4.0 \times \text{X}_{\text{Ca}}$	+0.0 to $+0.4%$	no correction
high-Ca pyroxene (En ₅₄₋₇₄ Wo ₂₆₋₄₅)	$\Delta \delta^7 \text{Li}_{\text{en-sl1}}^{\text{phase}} = 4.0 \times \text{X}_{\text{Ca}}$	+1.0 to +1.8%	no correction
plagioclase ($Ab_{17-19}An_{81-83}$)	$\Delta \delta^7 \text{Li}_{\text{ol-sc1}}^{\text{phase}} = -0.02 \times X_{\text{Na}} - 0.63$	-0.6 to $-0.6%$	no correction

Table S4: Spot-to-spot O- and Li-isotope compositions of chondrule constituents. Uncertainty (1SD) is shown in parenthesis.

		phase	occurrence	compo*	O-analysis ID	Li-analysis ID	δ^{18} O	δ^{17} O	$\Delta^{17}O$	δ^7 Li	$[Li]^{\mu g \cdot g^{-1}}$
group 1	BOP-2	Ol	shell of inner BO	Fo ₉₅	oxy-ol-chd2@9364	liso-ol-chd2@8692	-4.1 (1)	-7.2 (2)	-5.1	8.2 (1.3)	0.153
	BOP-2	Ol	shell of inner BO	F094	oxy-ol-chd2@9365	liso-ol-chd2@8693	-4.7 (1)	-7.9 (3)	-5.5	4.2 (0.79)	1.42
	BOP-2	Ol	shell of inner BO	Fo ₉₆	oxy-ol-chd2@9366	liso-ol-chd2@8694	-3.2 (1)	-5.8 (2)	-4.1	8.5 (1.1)	0.289
	BOP-2	Ol	shell of inner BO	Fo ₉₅	oxy-ol-chd2@9433	liso-ol-chd2@8671	-3.9 (2)	-7.4 (3)	-5.4	15.2 (0.63)	0.693
	BOP-2	Ol	shell of inner BO	Fo ₉₈	oxy-ol-chd2@9434	liso-ol-chd2@8672	-3.9 (1)	-7.7 (3)	-5.7	10.4 (1.2)	0.19:
	BOP-2	Ol	inner BO	F099	oxy-ol-chd2@9455	liso-ol-chd2@8705	-4.3 (1)	-7.9 (3)	-5.6	26.9 (2.0)	0.24
	BOP-2	Ol	shell of inner BO	Fo ₉₈	oxy-ol-chd2@9456	liso-ol-chd2@8706	-4.8 (1)	-8.0 (3)	-5.5	6.4 (1.2)	0.65
	BOP-2	Ol	inner BO	F099	oxy-ol-chd2@9473	liso-ol-chd2@8709	-5.8 (1)	-8.4 (3)	-5.4	23.8 (1.1)	0.71
	BOP-2	Ol	inner BO	Fo ₉₈	oxy-ol-chd2@9474	liso-ol-chd2@8674	-4.6 (1)	-8.3 (3)	-5.9	27.1 (1.3)	0.56
	BOP-2	Ol	shell of outer BOP	Fo ₉₈	oxy-ol-chd2@9492	liso-ol-chd2@8945	-4.7 (1)	-8.3 (3)	-5.9	12.9 (1.9)	0.2
	BOP-2	Ol	shell of outer BOP	Fo ₉₇	oxy-ol-chd2@9493	liso-ol-chd2@8946	-3.8 (1)	-7.5 (2)	-5.6	16.3 (3.0)	0.13
	BOP-2	Ol	shell of outer BOP	Fo ₉₈	oxy-ol-chd2@9494	liso-ol-chd2@8947	-3.8 (2)	-7.8 (4)	-5.8	4.7 (3.2)	0.081
	BOP-2	Ol	shell of inner BO	Fo ₉₈	oxy-ol-chd2@9504	liso-ol-chd2@8712	-4.4 (1)	-7.4 (3)	-5.1	-0.6 (1.0)	0.87
	BOP-2	Ol	inner BO	F099	oxy-ol-chd2@9505	liso-ol-chd2@8726	-3.3 (1)	-7.0 (3)	-5.2	11.7 (1.1)	0.71
	BOP-2	Ol	shell of inner BO	Fo ₉₆	oxy-ol-chd2@9507	liso-ol-chd2@8975	-6.3 (2)	-8.4 (4)	-5.1	9.9 (1.2)	0.88
	BOP-2	Ol	shell of outer BOP	Fo ₉₇	oxy-ol-chd2@9509	liso-ol-chd2@8977	-7.0(2)	-8.4 (3)	-4.7	13.6 (1.8)	0.23
	BOP-2	Ol	shell of outer BOP	Fo ₉₆	oxy-ol-chd2@9511	liso-ol-chd2@8979	-5.5 (1)	-7.9 (3)	-5.1	7.3 (1.5)	0.37
	BOP-2	Ol	shell of outer BOP	Fo ₉₅	oxy-ol-chd2@9512	liso-ol-chd2@8980	-6.0(1)	-8.4 (3)	-5.2	-5.0 (1.9)	0.21
	BOP-2	Ol	shell of outer BOP	F099	oxy-ol-chd2@9513	liso-ol-chd2@8981	-5.5 (1)	-7.7 (3)	-4.9	2.2 (1.4)	0.46
	BOP-2	Ol	shell of outer BOP	Fo ₉₆	oxy-ol-chd2@9515	liso-ol-chd2@8948	-5.0 (1)	-7.7 (3)	-5.1	5.6 (2.9)	0.12
	BOP-2	Ol	shell of outer BOP	Fo ₉₆	oxy-ol-chd2@9733	liso-ol-chd2@8949	-3.4(1)	-7.2 (3)	-5.4	7.7 (2.4)	0.16
	BOP-2	Ol	shell of outer BOP	Fo ₉₈	oxy-ol-chd2@9734	liso-ol-chd2@8950	-4.6(1)	-7.5 (3)	-5.1	9.3 (2.8)	0.06
	BOP-2	Ol	shell of outer BOP	Fo ₉₈	oxy-ol-chd2@9735	liso-ol-chd2@8951	-3.9(2)	-7.0 (3)	-4.9	8.2 (3.3)	0.094
	BOP-2	Ol	outer BOP	Fo ₉₇	oxy-ol-chd2@9737	liso-ol-chd2@8952	-4.5 (2)	-7.7 (3)	-5.4	7.0 (2.4)	0.19
	BOP-2	Ol	outer BOP	Fo ₉₈	oxy-ol-chd2@9738	liso-ol-chd2@8953	-4.7 (1)	-8.2 (3)	-5.7	26.2 (1.8)	0.30
	BOP-2	Ol	shell of outer BOP	Fo ₉₆	oxy-ol-chd2@9739	liso-ol-chd2@8954	-4.3(1)	-8.0 (3)	-5.7	9.0 (2.3)	0.13
	BOP-2	Ol	shell of inner BO	F099	oxy-ol-chd2@9749	liso-ol-chd2@8673	-5.4(2)	-9.0(3)	-6.2	7.1 (0.91)	0.34
	BOP-2	Ol	shell of inner BO	Fo ₉₈	oxy-ol-chd2@9750	liso-ol-chd2@8710	-3.9(2)	-8.0 (3)	-6.0	9.0 (1.5)	0.43
	BOP-2	Ol	shell of inner BO	Fo ₉₆	oxy-ol-chd2@9751	liso-ol-chd2@8711	-5.2(1)	-8.2 (3)	-5.5	4.6 (1.2)	0.84
	BOP-2	Ol	shell of inner BO	F099	oxy-ol-chd2@9753	liso-ol-chd2@8713	-4.3 (2)	-7.2 (3)	-5.0	5.3 (1.2)	0.82
	BOP-2	Ol	shell of inner BO	Fo ₈₈	oxy-ol-chd2@9754	liso-ol-chd2@8714	-5.6(1)	-8.4 (4)	-5.4	6.6 (1.4)	0.49
	BOP-2	Ol	shell of inner BO	F099	oxy-ol-chd2@9758	liso-ol-chd2@8715	-4.1(1)	-7.6 (4)	-5.4	3.9 (1.0)	0.74
	BOP-2	Ol	shell of inner BO	Fo ₉₉	oxy-ol-chd2@9759	liso-ol-chd2@8716	-4.3 (1)	-7.5 (4)	-5.3	7.2 (1.3)	0.58
	BOP-2	Ol	shell of inner BO	F099	oxy-ol-chd2@9760	liso-ol-chd2@8717	-5.7 (1)	-8.1 (3)	-5.2	3.0 (2.7)	0.14
	BOP-2	Ol	inner BO	F098	oxy-ol-chd2@9762	liso-ol-chd2@8727	-3.3 (2)	-6.5 (3)	-4.7	26.8 (2.0)	0.22
	BOP-2	Ol	shell of inner BO	Fo ₉₈	oxy-ol-chd2@9763	liso-ol-chd2@8728	-5.1 (1)	-7.8 (4)	-5.1	7.6 (0.96)	1.2
	BOP-2	Ol	shell of inner BO	Fo ₉₉	oxy-ol-chd2@9764	liso-ol-chd2@8729	-4.2(1)	-8.1 (3)	-5.9	0.9 (1.9)	0.35
	BOP-2	Ol	shell of inner BO	F097	oxy-ol-chd2@9868	liso-ol-chd2@8736	-6.6 (1)	-8.7 (3)	-5.3	3.7 (1.4)	0.43
	BOP-2	Ol	shell of inner BO	Fo ₉₈	oxy-ol-chd2@9869	liso-ol-chd2@8737	-6.5 (1)	-7.8 (3)	-4.4	8.6 (1.2)	0.64

Table S4: Continued

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	phase	occurrence	compo*	O-analysis ID	Li-analysis ID	δ^{18} O	$\delta^{17}O$	$\Delta^{17}O$	δ^7 Li	$[Li]^{\mu g \cdot g^{-1}}$
BOP-2	Ol	shell of inner BO	F099	oxy-ol-chd2@9870	liso-ol-chd2@8738	-5.4 (2)	-8.4 (3)	-5.5	3.3 (1.4)	0.795
BOP-2	O1	shell of inner BO	F099	oxy-ol-chd2@9872	liso-ol-chd2@8739	-4.4 (2)	-7.9 (3)	-5.6	12.7 (2.1)	0.342
BOP-2	Ol	shell of inner BO	F099	oxy-ol-chd2@9873	liso-ol-chd2@8740	-5.3 (2)	-8.7 (3)	-6.0	6.8 (1.8)	0.28
BOP-2	Ol	shell of inner BO	F099	oxy-ol-chd2@9874	liso-ol-chd2@8741	-5.1 (2)	-8.2 (4)	-5.5	7.0 (2.3)	0.355
BOP-2	Ol	shell of inner BO	Fo ₉₇	oxy-ol-chd2@9876	liso-ol-chd2@8742	-5.3 (2)	-8.4 (3)	-5.7	17.0 (1.8)	0.367
BOP-2	Ol	shell of inner BO	Fo ₉₈	oxy-ol-chd2@9877	liso-ol-chd2@8743	-6.1 (1)	-8.1 (3)	-4.9	7.8 (1.5)	0.437
BOP-2	Ol	inner BO	F099	oxy-ol-chd2@9879	liso-ol-chd2@8707	-5.2 (2)	-8.3 (3)	-5.5	15.4 (1.7)	0.225
BOP-2	Ol	inner BO	F099	oxy-ol-chd2@9880	liso-ol-chd2@8708	-5.4 (1)	-7.9 (3)	-5.1	19.9 (1.2)	0.599
BOP-2	Ol	shell of outer BOP	Fo ₉₆	oxy-ol-chd2@9881	liso-ol-chd2@8978	-5.6 (1)	-8.7 (3)	-5.8	9.5 (1.1)	0.637
BOP-2	Ol	shell of inner BO	Fo ₉₅	oxy-ol-chd2@9892	liso-ol-chd2@8973	-4.3 (2)	-7.5 (3)	-5.2	6.9 (1.2)	0.561
BOP-2	Ol	shell of inner BO	Fo ₉₇	oxy-ol-chd2@9893	liso-ol-chd2@8974	-5.0(1)	-7.4 (4)	-4.8	5.6 (1.1)	0.186
BOP-2	Ol	shell of inner BO	Fo ₉₈	oxy-ol-chd2@9896	liso-ol-chd2@8696	-5.0(1)	-8.0(2)	-5.5	11.5 (1.3)	0.44
BOP-2	Ol	shell of inner BO	Fo ₉₈	oxy-ol-chd2@9897	liso-ol-chd2@8695	-4.9(1)	-7.6 (2)	-5.0	11.5 (1.5)	0.547
BOP-2	Olirim	igneous rim	Fo ₈₉	oxy-ol-chd2@9902	liso-ol-chd2@8982	-4.7(1)	-7.7 (3)	-5.2	4.3 (1.7)	0.264
BOP-2	Olirim	igneous rim	Fo ₉₂	oxy-ol-chd2@9903	liso-ol-chd2@8983	-4.9(2)	-8.1 (3)	-5.5	5.7 (2.4)	0.125
BOP-2	Ol	shell of outer BOP	Fo ₉₈	oxy-ol-chd2@9920	liso-ol-chd2@8955	-4.3 (2)	-7.6 (3)	-5.4	12.6 (2.1)	0.193
BOP-2	Ol	shell of outer BOP	Fo ₉₈	oxy-ol-chd2@9921	liso-ol-chd2@8956	-4.3 (2)	-7.8 (3)	-5.5	15.2 (1.9)	0.216
BOP-2	Ol	shell of outer BOP	Fo ₉₇	oxy-ol-chd2@9923	liso-ol-chd2@8957	-4.0(2)	-7.2 (4)	-5.1	7.3 (3.0)	0.361
BOP-2	Ol	shell of outer BOP	Fo ₉₆	oxy-ol-chd2@9924	liso-ol-chd2@8958	-4.3 (2)	-7.0 (3)	-4.8	4.6 (2.2)	0.227
BOP-2	Ol	shell of outer BOP	Fo ₉₈	oxy-ol-chd2@9925	liso-ol-chd2@8959	-3.9(1)	-7.4 (3)	-5.4	10.0 (2.1)	0.188
BOP-2	Ol	shell of inner BO	Fo ₉₄	oxy-ol-chd2@10013	liso-ol-chd2@8730	-4.8(1)	-8.5 (4)	-5.9	4.1 (1.4)	0.666
BOP-2	Ol	shell of inner BO	Fo ₉₈	oxy-ol-chd2@10014	liso-ol-chd2@8731	-4.5 (2)	-7.9 (3)	-5.6	4.6 (1.3)	0.713
BOP-2	Ol	shell of inner BO	Fo ₉₆	oxy-ol-chd2@10015	liso-ol-chd2@8732	-5.1 (2)	-7.4(2)	-4.8	4.2 (0.74)	1.74
BOP-2	Ol	shell of inner BO	F099	oxy-ol-chd2@10017	liso-ol-chd2@8733	-4.8 (2)	-8.3 (3)	-5.9	4.4 (1.8)	0.304
BOP-2	Ol	shell of inner BO	F099	oxy-ol-chd2@10018	liso-ol-chd2@8734	-5.0(1)	-7.9 (3)	-5.3	4.0 (0.94)	1.38
BOP-2	Ol	shell of inner BO	F099	oxy-ol-chd2@10019	liso-ol-chd2@8735	-5.1 (2)	-7.8 (3)	-5.1	5.1 (0.94)	1
BOP-2	Ol	shell of inner BO	Fo ₉₉	oxy-ol-chd2@10021	liso-ol-chd2@8968	-5.2 (2)	-7.8 (3)	-5.1	2.0 (1.3)	1.49
BOP-2	Ol	shell of inner BO	Fo ₉₁	oxy-ol-chd2@10022	liso-ol-chd2@8969	-5.2 (2)	-8.1 (3)	-5.4	3.6 (1.4)	1.1
BOP-2	Ol	shell of inner BO	F099	oxy-ol-chd2@10023	liso-ol-chd2@8970	-5.4(1)	-8.1 (3)	-5.3	0.4(1.8)	0.826
BOP-2	Ol	shell of inner BO	Fo ₉₈	oxy-ol-chd2@10025	liso-ol-chd2@8971	-5.2(1)	-8.4 (4)	-5.7	-3.5 (1.0)	1.58
BOP-2	Ol	shell of inner BO	Fo ₉₃	oxy-ol-chd2@10026	liso-ol-chd2@8972	-5.2(1)	-7.6 (3)	-4.9	4.3 (1.3)	0.944
BOP-2	LPxirim	igneous rim	En ₉₈ Wo ₁	oxy-opx-chd2@9371	liso-opx-chd2@8787	-2.3(1)	-6.1 (3)	-4.9	-9.5 (1.5)	5.67
BOP-2	LPxirim	igneous rim	En ₉₈ Wo ₁	oxy-opx-chd2@9372	liso-opx-chd2@8788	-0.5 (1)	-4.6 (3)	-4.4	-8.2 (1.5)	5.54
BOP-2	LPx	outer BOP	En ₉₈ Wo ₁	oxy-opx-chd2@9436	liso-opx-chd2@8824	-3.1 (1)	-7.4 (4)	-5.8	10.5 (2.0)	1.28
BOP-2	LPx	outer BOP	En ₉₈ Wo ₁	oxy-opx-chd2@9437	liso-opx-chd2@8826	-2.7(1)	-7.8 (3)	-6.4	33.2 (3.8)	0.405
BOP-2	LPx	outer BOP	En ₉₈ Wo ₂	oxy-opx-chd2@9438	liso-opx-chd2@8827	-3.3 (2)	-7.4 (3)	-5.7	1.5 (1.1)	4.75
BOP-2	LPx	outer BOP	En ₉₈ Wo ₁	oxy-opx-chd2@9440	liso-opx-chd2@8819	-3.3 (1)	-6.8 (3)	-5.1	-33.2 (1.2)	2.75
BOP-2	LPx	outer BOP	En ₉₉ Wo ₁	oxy-opx-chd2@9441	liso-opx-chd2@8830	-2.4(1)	-6.5 (3)	-5.3	-8.0 (1.1)	5.03
BOP-2	LPx	outer BOP	En ₉₈ Wo ₁	oxy-opx-chd2@9442	liso-opx-chd2@8821	-3.0 (2)	-7.3 (3)	-5.7	-9.2 (1.7)	1.68
BOP-2	LPx ^{irim}	igneous rim	En ₉₈ Wo ₁	oxy-opx-chd2@9444	liso-opx-chd2@8856	-3.1 (2)	-7.3 (3)	-5.7	0.0 (1.4)	4.64
BOP-2	LPx ^{irim}	igneous rim	En ₉₉ Wo ₁	oxy-opx-chd2@9445	liso-opx-chd2@8857	-3.5 (1)	-7.4 (3)	-5.6	-0.6 (1.5)	5.55
DOI -2	LIA	15110003 11111	1199 1101	ony oph chaze 7443	1150 Opx Cliu2 @ 0057	3.3 (1)	7.4 (3)	5.0	0.0 (1.5)	5.55

Table S4: Continued

	phase	occurrence	compo*	O-analysis ID	Li-analysis ID	$\delta^{18}{ m O}$	$\delta^{17}{ m O}$	$\Delta^{17}O$	δ^7 Li	$[Li]^{\mu g \cdot g^{-1}}$
BOP-2	LPx	outer BOP	En ₉₉ Wo ₁	oxy-opx-chd2@9487	liso-opx-chd2@8785	-3.8 (2)	-8.3 (3)	-6.3	-10.3 (0.61)	5.92
BOP-2	LPx	outer BOP	$En_{99}Wo_1$	oxy-opx-chd2@9488	liso-opx-chd2@8786	-2.6 (1)	-7.0 (4)	-5.6	-14.2 (0.86)	3.19
BOP-2	LPx	outer BOP	$En_{98}Wo_1$	oxy-opx-chd2@9489	liso-opx-chd2@8809	-2.5 (2)	-6.6 (3)	-5.3	-7.1 (2.5)	1.13
BOP-2	LPx ^{irim}	igneous rim	$En_{98}Wo_1$	oxy-opx-chd2@9894	liso-opx-chd2@8925	-3.1(1)	-6.5 (3)	-4.9	0.0 (1.6)	4.82
BOP-2	LPx	outer BOP	$En_{98}Wo_1$	oxy-opx-chd2@9899	liso-opx-chd2@8915	-2.3(2)	-6.9 (3)	-5.7	-4.0 (1.5)	2.96
BOP-2	LPx	outer BOP	$En_{98}Wo_1$	oxy-opx-chd2@9900	liso-opx-chd2@8916	-3.0(2)	-6.9 (4)	-5.3	-6.0 (1.3)	4.44
BOP-2	LPxirim	igneous rim	$En_{98}Wo_1$	oxy-opx-chd2@9904	liso-opx-chd2@8858	-3.0(1)	-6.5 (3)	-4.9	-10.9 (1.6)	4.72
BOP-2	LPxirim	igneous rim	$En_{98}Wo_1$	oxy-opx-chd2@9905	liso-opx-chd2@8926	-1.2(1)	-5.9 (4)	-5.2	0.3 (1.3)	5.23
BOP-2	LPx	outer BOP	$En_{97}Wo_2$	oxy-opx-chd2@9936	liso-opx-chd2@8829	-3.4(1)	-7.4 (3)	-5.6	-10.1 (0.76)	5.92
BOP-2	LPx	outer BOP	$En_{98}Wo_1$	oxy-opx-chd2@9938	liso-opx-chd2@8823	-3.7 (2)	-7.8 (4)	-5.8	-12.9 (1.1)	3.18
BOP-2	LPx	outer BOP	$En_{98}Wo_1$	oxy-opx-chd2@9940	liso-opx-chd2@8843	-3.2(1)	-7.3 (3)	-5.6	20.3 (2.4)	0.899
BOP-2	LPx	outer BOP	En ₉₈ Wo ₁	oxy-opx-chd2@9941	liso-opx-chd2@8825	-3.0(1)	-6.8 (3)	-5.2	24.8 (2.6)	0.658
BOP-2	LPx	outer BOP	$En_{97}Wo_2$	oxy-opx-chd2@9942	liso-opx-chd2@8828	-2.6(1)	-6.6 (3)	-5.3	-5.8 (1.1)	3.64
BOP-2	LPx	outer BOP	En ₉₈ Wo ₁	oxy-opx-chd2@9944	liso-opx-chd2@8820	-2.4(2)	-6.8 (3)	-5.6	-0.4 (1.4)	4.75
BOP-2	LPx	outer BOP	$En_{99}Wo_1$	oxy-opx-chd2@9945	liso-opx-chd2@8831	-1.8 (2)	-6.1 (3)	-5.1	-20.8 (1.1)	2.46
BOP-2	LPx	outer BOP	$En_{97}Wo_2$	oxy-opx-chd2@9946	liso-opx-chd2@8847	-2.5(1)	-7.2 (3)	-5.9	-0.7 (1.1)	4.96
BOP-2	LPx	outer BOP	En ₉₈ Wo ₁	oxy-opx-chd2@9948	liso-opx-chd2@8848	-2.1(2)	-6.4 (3)	-5.3	-21.5 (1.6)	2.78
BOP-2	LPx	outer BOP	$En_{99}Wo_1$	oxy-opx-chd2@9949	liso-opx-chd2@8849	-1.6(2)	-6.1 (3)	-5.3	-4.1 (1.2)	5.25
BOP-2	LPx	outer BOP	$En_{99}Wo_1$	oxy-opx-chd2@9950	liso-opx-chd2@8850	-2.5(1)	-6.6 (3)	-5.3	-13.0 (1.3)	2.92
BOP-2	LPx	outer BOP	En ₉₈ Wo ₁	oxy-opx-chd2@9952	liso-opx-chd2@8851	-2.8(1)	-6.6 (3)	-5.1	-23.2 (1.4)	2.64
BOP-2	LPx	outer BOP	En ₉₈ Wo ₁	oxy-opx-chd2@9953	liso-opx-chd2@8852	-2.3 (1)	-6.6 (3)	-5.4	-17.1 (1.0)	4.37
BOP-2	LPx	outer BOP	$En_{97}Wo_2$	oxy-opx-chd2@9964	liso-opx-chd2@8893	-3.8 (2)	-7.5 (3)	-5.5	-2.6 (0.82)	6.73
BOP-2	LPx	outer BOP	$En_{98}Wo_1$	oxy-opx-chd2@9965	liso-opx-chd2@8894	-4.1(2)	-7.3 (4)	-5.2	-10.8 (1.1)	5.27
BOP-2	LPx	outer BOP	$En_{97}Wo_2$	oxy-opx-chd2@9966	liso-opx-chd2@8895	-3.8(1)	-7.4 (3)	-5.4	-12.2 (1.1)	4.57
BOP-2	LPx	outer BOP	En ₉₈ Wo ₁	oxy-opx-chd2@9968	liso-opx-chd2@8896	-4.7 (2)	-7.9 (3)	-5.4	-4.0 (1.0)	6.35
BOP-2	LPx	outer BOP	En ₉₈ Wo ₁	oxy-opx-chd2@9969	liso-opx-chd2@8897	-3.8(1)	-6.6 (3)	-4.6	0.2(1.1)	3.64
BOP-2	LPx	outer BOP	$En_{96}Wo_3$	oxy-opx-chd2@9972	liso-opx-chd2@8899	-3.4(2)	-7.2 (3)	-5.5	19.4 (2.3)	0.253
BOP-2	LPx	outer BOP	En ₉₄ Wo ₅	oxy-opx-chd2@9973	liso-opx-chd2@8900	-3.2 (2)	-7.0 (3)	-5.3	1.8 (1.5)	1.56
BOP-2	LPx	outer BOP	En ₉₆ Wo ₄	oxy-opx-chd2@9974	liso-opx-chd2@8901	-3.3 (2)	-7.3 (3)	-5.6	10.3 (2.0)	0.258
BOP-2	LPx	outer BOP	$En_{95}Wo_4$	oxy-opx-chd2@9986	liso-opx-chd2@8902	-3.8 (2)	-7.7 (3)	-5.7	-3.4 (1.7)	1.64
BOP-2	LPx	outer BOP	$En_{94}Wo_5$	oxy-opx-chd2@9987	liso-opx-chd2@8911	-3.8 (2)	-7.4 (3)	-5.4	-0.4 (1.7)	0.906
BOP-2	LPx	outer BOP	En ₉₃ Wo ₆	oxy-opx-chd2@9988	liso-opx-chd2@8912	-3.2(1)	-6.0 (4)	-4.4	-2.9 (1.8)	1.47
BOP-2	LPx	outer BOP	$En_{91}Wo_8$	oxy-opx-chd2@9990	liso-opx-chd2@8913	-2.1(2)	-5.8 (3)	-4.7	-1.6 (1.5)	1.35
BOP-2	LPx	outer BOP	En ₉₀ Wo ₉	oxy-opx-chd2@9991	liso-opx-chd2@8914	-4.0(1)	-6.8 (3)	-4.8	-1.1 (1.8)	1.23
BOP-2	LPx	outer BOP	$En_{99}Wo_1$	oxy-opx-chd2@9992	liso-opx-chd2@8808	-2.8 (2)	-5.7 (4)	-4.3	-7.3 (2.1)	2.07
BOP-2	LPx	outer BOP	$En_{99}Wo_1$	oxy-opx-chd2@9994	liso-opx-chd2@8810	-3.0(1)	-7.3 (3)	-5.8	0.3 (2.3)	2.26
BOP-2	LPx	outer BOP	En ₉₈ Wo ₁	oxy-opx-chd2@9995	liso-opx-chd2@8844	-3.9 (2)	-7.9 (4)	-5.8	-2.6 (1.1)	4.47
BOP-2	LPx	outer BOP	En ₉₈ Wo ₁	oxy-opx-chd2@9996	liso-opx-chd2@8846	-3.0 (1)	-7.1 (3)	-5.6	-7.5 (0.86)	4.7
BOP-2	HPx	outer BOP	En ₇₂ Wo ₂₇	oxy-cpx-chd2@9462	liso-cpx-chd2@8917	-3.5 (1)	-8.0 (3)	-6.2	0.5 (2.5)	0.324
BOP-2	HPx	outer BOP	En ₇₁ Wo ₂₈	oxy-cpx-chd2@9463	liso-cpx-chd2@8918	-1.1 (2)	-5.3 (3)	-4.7	-8.8 (2.7)	0.506
BOP-2	HPx	outer BOP	En ₆₅ Wo ₃₅	oxy-cpx-chd2@9464	liso-cpx-chd2@8919	-1.6 (1)	-6.8 (3)	-5.9	-4.9 (2.6)	0.435
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Table S4: Continued

	mhasa			O analysis ID	Li analysis ID	δ^{18} O	δ^{17} O	Δ ¹⁷ O	δ^7 Li	[Li] ^{\(\mu g \cdot g^{-1}\)}
BOP-2	phase HPx	occurrence outer BOP	compo* En ₆₉ Wo ₃₁	O-analysis ID oxy-cpx-chd2@9483	Li-analysis ID liso-cpx-chd2@8853	-2.7 (1)	-6.9 (3)	-5.5	-14.1 (3.3)	0.388
BOP-2 BOP-2	HPx	outer BOP	En ₆₉ Wo ₃₁ En ₅₇ Wo ₄₂	oxy-cpx-chd2@9484	liso-cpx-chd2@8854	-2.7 (1) -2.5 (1)	-6.9 (3) -6.7 (3)	-5.3 -5.4	0.8 (2.7)	1.06
BOP-2	HPx	outer BOP	En ₅₇ Wo ₄₂ En ₇₄ Wo ₂₆	oxy-cpx-chd2@9485	liso-cpx-chd2@8920	-2.3 (1)	-0.7 (3) -7.8 (4)	-5.4 -6.1	-6.8 (2.2)	0.747
BOP-2 BOP-2	ri Pl	inner BO	An ₈₁ Ab ₁₉	oxy-cpx-chd2@9488	liso-pl-chd2@9206	-3.2 (1) 4.1 (1)	-7.8 (4) -1.1 (3)	-3.2	6.4 (5.6)	0.0079
BOP-2 BOP-2	Pl	inner BO		oxy-pl-chd2@9459		3.9 (1)	-1.1 (3) -1.6 (3)	-3.2 -3.7	-4.6 (1.4)	0.0079
BOP-2 BOP-2	Pl	inner BO	$An_{83}Ab_{17} An_{81}Ab_{19}$		liso-pl-chd2@9212 liso-pl-chd2@9213	3.9 (1) 4.6 (1)	-0.3 (3)	-3.7 -2.7	1.3 (2.6)	0.129
BOP-2 BOP-2	Pl	outer BOP	01 17	oxy-pl-chd2@9460	*		` '	-2.7	, ,	0.0403
BOP-2 BOP-2	Pl Pl	outer BOP	An ₈₃ Ab ₁₇	oxy-pl-chd2@9476 oxy-pl-chd2@9477	liso-pl-chd2@9215 liso-pl-chd2@9232	6.0 (1) 6.3 (1)	0.0 (3) -0.2 (3)	-3.1 -3.4	-2.0 (2.9) -2.0 (5.6)	0.0224
BOP-2 BOP-2	Pl	outer BOP	An ₈₂ Ab ₁₈	* I	1	` '	` '	-3.4	` /	0.00972
BOP-2 BOP-2	Pl	inner BO	An ₈₂ Ab ₁₈	oxy-pl-chd2@9478	liso-pl-chd2@9233	6.8 (1)	0.2 (2)	-3.3 -3.7	1.5 (2.1) 10.3 (5.6)	0.0324
BOP-2 BOP-2	Pl	inner BO	An ₈₁ Ab ₁₉	oxy-pl-chd2@9480	liso-pl-chd2@9235	4.9 (2)	-1.1 (3)	-3.7	` /	
	PI Pl	inner BO	$An_{81}Ab_{18}$	oxy-pl-chd2@9481	liso-pl-chd2@9236	4.9 (2)	-0.6 (3)		0.4 (6.4)	0.0257
BOP-2 BOP-2	Pl Pl	outer BOP	An ₈₃ Ab ₁₇	oxy-pl-chd2@10010	liso-pl-chd2@9214	2.1 (1)	-3.4 (4)	-4.5 -2.7	-1.3 (2.9)	0.0356
			$An_{82}Ab_{18}$	oxy-pl-chd2@10011	liso-pl-chd2@9234	7.0 (1)	0.9 (3)		2.9 (2.4)	0.0525
PO-6	Ol	main	Fo ₉₉	oxy-ol-chd6@9774	liso-ol-chd6@9115	-7.0 (1)	-10.1 (3)	-6.5	-8.4 (1.2)	0.504
PO-6	Ol Ol ^{FeO}	main	F094	oxy-ol-chd6@9775	liso-ol-chd6@9116	-4.6 (1)	-6.9 (3)	-4.5	6.1 (1.4)	0.31
PO-6		main	Fo ₆₅	oxy-ol-chd6@9776	liso-ol-chd6@9117	4.9 (2)	1.0 (3)	-1.6	3.4 (2.8)	0.274
PO-6	Ol	main	F098	oxy-ol-chd6@9778	liso-ol-chd6@9118	-5.1 (2)	-7.4 (2)	-4.7	2.0 (1.3)	0.855
PO-6	Ol	main	Fo ₈₆	oxy-ol-chd6@9781	liso-ol-chd6@9120	-3.6 (1)	-6.9 (3)	-5.1	1.9 (2.4)	0.0836
PO-6	Ol	main	Fo ₈₈	oxy-ol-chd6@9782	liso-ol-chd6@9121	-4.8 (1)	-7.6 (4)	-5.1	3.9 (3.1)	0.0882
PO-6	Ol	main	Fo ₈₈	oxy-ol-chd6@9783	liso-ol-chd6@9122	-3.7 (2)	-7.2 (3)	-5.3	7.7 (2.7)	0.115
PO-6	Ol ^{irim}	igneous rim	Fo ₉₀	oxy-ol-chd6@9793	liso-ol-chd6@9124	-4.6 (2)	-7.2 (3)	-4.8	9.0 (1.7)	0.26
PO-6	Ol ^{irim}	igneous rim	Fo ₈₅	oxy-ol-chd6@9794	liso-ol-chd6@9125	-4.5 (2)	-7.2 (3)	-4.9	-0.9 (4.2)	0.0532
PO-6	Ol ^{irim}	igneous rim	Fo ₇₈	oxy-ol-chd6@9796	liso-ol-chd6@9126	-4.0 (1)	-6.5 (3)	-4.4	8.9 (5.6)	0.0435
PO-6	Ol ^{FeO}	main	Fo ₆₃	oxy-ol-chd6@9805	liso-ol-chd6@9127	4.6 (1)	-1.2 (4)	-3.6	0.9 (5.5)	0.0562
PO-6	LPxirim	igneous rim	$En_{96}Wo_1$	oxy-opx-chd6@9797	liso-opx-chd6@9175	-2.9 (2)	-6.0 (3)	-4.5	-14.8 (0.69)	9.01
PO-6	LPxirim	igneous rim	$En_{97}Wo_1$	oxy-opx-chd6@9798	liso-opx-chd6@9176	-3.8 (1)	-6.6 (4)	-4.6	-7.7 (0.75)	12.2
PO-6	LPxirim	igneous rim	$En_{98}Wo_1$	oxy-opx-chd6@9800	liso-opx-chd6@9173	-2.7(1)	-6.1 (3)	-4.7	-6.6 (0.78)	8.94
PO-6	LPxirim	igneous rim	$En_{98}Wo_1$	oxy-opx-chd6@9801	liso-opx-chd6@9174	-3.3 (2)	-7.1 (3)	-5.3	-17.1 (0.71)	4.85
PO-6	LPxirim	igneous rim	$En_{97}Wo_1$	oxy-opx-chd6@9803	liso-opx-chd6@9184	-3.6(2)	-7.2 (3)	-5.4	-21.2 (0.95)	7.05
PO-6	LPxirim	igneous rim	$En_{98}Wo_1$	oxy-opx-chd6@9804	liso-opx-chd6@9185	-4.5 (2)	-7.9 (3)	-5.6	-7.2 (0.73)	13.6
PO-6	LPxirim	igneous rim	$En_{98}Wo_1$	oxy-opx-chd6@9814	liso-opx-chd6@9186	-4.0(1)	-7.3 (4)	-5.2	-6.2 (1.0)	3.55
PO-6	LPxirim	igneous rim	En ₉₈ Wo ₁	oxy-opx-chd6@9815	liso-opx-chd6@9187	-3.4(2)	-6.9 (3)	-5.1	-14.3 (0.94)	10.5
PO-6	LPxirim	igneous rim	En ₉₈ Wo ₁	oxy-opx-chd6@9816	liso-opx-chd6@9188	-4.1 (1)	-7.3 (3)	-5.1	-14.5 (1.0)	5.52
PO-6	LPxirim	igneous rim	En ₉₈ Wo ₁	oxy-opx-chd6@9819	liso-opx-chd6@9189	-4.3 (2)	-7.4 (3)	-5.2	-25.0 (1.1)	5.01
PO-6	LPxirim	igneous rim	En ₉₈ Wo ₁	oxy-opx-chd6@9820	liso-opx-chd6@9190	-4.2 (1)	-7.2 (3)	-5.0	-21.7 (0.9)	5.61
PO-6	LPxirim	igneous rim	En ₉₈ Wo ₁	oxy-opx-chd6@9821	liso-opx-chd6@9191	-3.7 (2)	-6.7 (4)	-4.8	-17.8 (0.79)	6.7
BO-4	Ol	main	Fo ₉₈	oxx-ol-chd4@565	liso-ol-chd4@9019	-4.1 (3)	-7.9 (4)	-5.8	14.4 (2.7)	0.115
BO-4	Ol	main	F099	oxx-ol-chd4@566	liso-ol-chd4@9020	-4.1 (2)	-7.5 (4)	-5.4	6.6 (2.4)	0.106
BO-4	Ol	shell	F092	oxx-ol-chd4@567	liso-ol-chd4@9021	-4.9 (2)	-8.0 (4)	-5.5	16.7 (1.7)	0.287
					01 0110 . 0 / 021	(-)		0.0		

Table S4: Continued

		phase	occurrence	compo*	O-analysis ID	Li-analysis ID	δ^{18} O	δ^{17} O	$\Delta^{17}O$	δ^7 Li	$[Li]^{\mu g \cdot g^{-1}}$
	BO-4	Ol	shell	Fo ₉₀	oxy-ol-chd4@9833	liso-ol-chd4@9039	-4.8 (1)	-7.4 (3)	-4.9	-1.4 (4.5)	0.0588
	BO-4	Ol	main	Fo ₉₉	oxy-ol-chd4@9834	liso-ol-chd4@9040	-4.2 (2)	-7.1 (3)	-4.9	39.1 (2.7)	0.32
	BO-4	Ol	main	F099	oxy-ol-chd4@9837	liso-ol-chd4@9042	-4.3 (2)	-7.3 (2)	-5.0	-5.3 (4.0)	0.14
	BO-4	Ol	main	Fo ₉₈	oxy-ol-chd4@9838	liso-ol-chd4@9043	-5.5 (2)	-8.0 (4)	-5.1	5.9 (4.3)	0.0839
	BO-4	Ol	main	Fo ₉₈	oxy-ol-chd4@9841	liso-ol-chd4@9045	-5.4(2)	-9.2 (3)	-6.4	9.9 (2.0)	0.0836
	BO-4	Ol	main	Fo ₉₈	oxy-ol-chd4@9852	liso-ol-chd4@9046	-4.0 (2)	-7.8 (4)	-5.7	14.1 (1.6)	0.129
	BO-4	Ol	main	Fo ₉₈	oxy-ol-chd4@9854	liso-ol-chd4@9048	-3.6 (2)	-7.3 (3)	-5.4	2.7 (3.1)	0.0689
	BO-4	Ol	main	Fo ₉₇	oxy-ol-chd4@9856	liso-ol-chd4@9049	-4.7 (1)	-7.7 (3)	-5.3	6.1 (1.7)	0.16
	BO-4	Ol	main	Fo ₉₇	oxy-ol-chd4@9857	liso-ol-chd4@9022	-4.2(1)	-8.5 (3)	-6.3	18.9 (1.5)	0.20
group 2	PO-7	Ol	main	Fo ₉₈	oxy-ol-chd7@9571	liso-ol-chd7@9146	-6.1 (1)	-9.6 (3)	-6.4	0.1 (0.85)	1.5
	PO-7	Ol	main	Fo ₉₀	oxy-ol-chd7@9572	liso-ol-chd7@9147	-6.3 (1)	-9.3 (3)	-6.0	10.2 (1.1)	0.23
	PO-7	Ol	main	Fo ₉₈	oxy-ol-chd7@9582	liso-ol-chd7@9145	-5.6 (1)	-8.8 (3)	-5.9	-14.8 (1.1)	0.648
	PO-7	LPxirim	igneous rim	$En_{98}Wo_1$	oxy-opx-chd7@9575	liso-opx-chd7@9192	-5.4(2)	-8.6 (3)	-5.7	-15.0 (0.92)	8.52
	PO-7	LPxirim	igneous rim	$En_{98}Wo_1$	oxy-opx-chd7@9576	liso-opx-chd7@9193	-5.8 (1)	-9.4 (3)	-6.3	-9.5 (0.95)	7.8
	PO-7	LPxirim	igneous rim	En ₉₈ Wo ₁	oxy-opx-chd7@9578	liso-opx-chd7@9194	-5.8(1)	-9.5 (3)	-6.5	-14.4 (0.83)	6.8
	PO-1	Ol	shell-of-mantle	Fo99	oxy-ol-chd1@9674	liso-ol-chd1@9136	-7.1(1)	-10.0 (3)	-6.3	-6.2 (1.3)	0.49
	PO-1	Ol	shell-of-mantle	Fo ₉₃	oxy-ol-chd1@9675	liso-ol-chd1@9137	-6.9(1)	-9.3 (3)	-5.7	4.0 (1.3)	0.37
	PO-1	OlFeO	main	Fo ₆₅	oxy-ol-chd1@9676	liso-ol-chd1@9138	5.8(1)	0.8(3)	-2.2	-2.4 (4.8)	0.10
	PO-1	Ol	shell-of-mantle	Fo ₉₇	oxy-ol-chd1@9678	liso-ol-chd1@9139	-5.7 (2)	-9.6 (3)	-6.6	15.8 (4.5)	0.0
	PO-1	Ol	shell-of-mantle	Fo ₉₉	oxy-ol-chd1@9679	liso-ol-chd1@9140	-5.5 (2)	-9.9 (3)	-7.0	-5.0 (3.4)	0.069
	PO-1	Ol	shell-of-mantle	Fo ₁₀₀	oxy-ol-chd1@9680	liso-ol-chd1@9142	-6.4 (2)	-10.1 (3)	-6.8	13.0 (3.8)	0.082
	PO-1	LPx	main	En ₉₃ Wo ₆	oxy-opx-chd1@9682	liso-opx-chd1@9256	-5.1(2)	-8.8 (3)	-6.1	-5.9 (1.3)	3.5
	PO-1	LPx	main	En ₉₂ Wo ₄	oxy-opx-chd1@9683	liso-opx-chd1@9257	-5.5 (2)	-9.8 (3)	-6.9	-0.3 (1.4)	2.7
	PO-1	LPx	main	$En_{95}Wo_4$	oxy-opx-chd1@9684	liso-opx-chd1@9258	-5.0(1)	-8.6 (3)	-6.0	18.7 (1.9)	1.5
	PO-1	HPx	main	En ₅₆ Wo ₄₃	oxy-cpx-chd1@9686	liso-cpx-chd1@9259	-6.2(1)	-9.6 (3)	-6.4	8.7 (3.9)	0.12
	PO-1	HPx	main	En ₅₉ Wo ₄₀	oxy-cpx-chd1@9687	liso-cpx-chd1@9260	-5.8 (2)	-9.5 (3)	-6.5	11.3 (3.7)	0.50
	PO-1	HPx	main	$En_{54}Wo_{45}$	oxy-cpx-chd1@9688	liso-cpx-chd1@9261	-6.8 (2)	-9.7 (3)	-6.2	-6.1 (3.9)	0.48
	PO-3c	Ol	main	Fo ₉₃	oxy-ol-chd3c@9720	liso-ol-chd3c@9098	-5.9 (1)	-9.5 (3)	-6.4	-1.0 (3.5)	0.086
	PO-3c	Ol	main	Fo ₈₁	oxy-ol-chd3c@9721	liso-ol-chd3c@9099	-7.6(1)	-9.8 (2)	-5.9	-12.2 (3.6)	0.071
	PO-3c	Ol	main	Fo ₈₁	oxy-ol-chd3c@9723	liso-ol-chd3c@9100	-6.8(1)	-10.2 (2)	-6.7	-4.9 (3.4)	0.078
	PO-3c	OlFeO	main	Fo ₆₂	oxy-ol-chd3c@9724	liso-ol-chd3c@9101	-2.7(2)	-4.5 (2)	-3.1	5.0 (4.9)	0.048
	PO-3s	Ol	main	Fo ₉₉	oxy-ol-chd3s@9726	liso-ol-chd3s@9102	-6.8 (1)	-10.0 (3)	-6.4	-4.6 (1.6)	0.39
	PO-3s	Ol	main	Fo ₈₆	oxy-ol-chd3s@9727	liso-ol-chd3s@9103	-7.1(1)	-9.7 (3)	-6.0	5.6 (1.1)	0.70
	PO-3s	Ol	main	Fo ₉₄	oxy-ol-chd3s@9729	liso-ol-chd3s@9104	-6.9(1)	-8.8 (2)	-5.3	5.6 (1.8)	0.54
	PO-3s	Ol	main	Fo ₉₇	oxy-ol-chd3s@9730	liso-ol-chd3s@9105	-6.4(2)	-9.5 (3)	-6.2	-18.2 (1.7)	0.30
	PO-3s	Ol	main	Fo ₉₄	oxy-ol-chd3s@9731	liso-ol-chd3s@9106	-6.2(1)	-9.1 (4)	-5.8	-5.6 (2.2)	0.35
group 3	PO-8	Ol	mantle	Fo ₉₆	oxx-ol-chd8@601	lixo-ol-chd8@601	-16.7 (2)	-17.8 (3)	-9.1	-1.9 (1.1)	0.29
J 1	PO-8	Ol	mantle	Fo ₉₃	oxx-ol-chd8@602	lixo-ol-chd8@602	-44.2 (2)	-44.4 (5)	-21.4	-18.0 (1.1)	0.95
	PO-8	Ol	mantle	Fo ₉₂	oxx-ol-chd8@603	lixo-ol-chd8@603	-45.4 (2)	-46.4 (3)	-22.8	-6.0 (1.4)	0.20
	PO-8	Ol	mantle	F097	oxx-ol-chd8@745	lixo-ol-chd8@745	-49.9 (2)	-47.9 (4)	-22.0	-14.6 (1.6)	0.14
	PO-8	Ol	mantle	Fo ₉₆	oxx-ol-chd8@746	lixo-ol-chd8@746	-48.9 (2)	-48.7 (4)	-23.3	-7.7 (1.6)	0.12

Table S4: Continued

	phase	occurrence	compo*	O-analysis ID	Li-analysis ID	δ^{18} O	$\delta^{17}O$	Δ ¹⁷ O	δ^7 Li	[Li] ^{µg·g-1}
PO-8	Ol ^{irim}	igneous rim	Fo ₈₄	oxx-ol-chd8@747	lixo-ol-chd8@747	-20.8 (2)	-24.3 (4)	-13.5	-3.6 (2.0)	0.0939
PO-8	Ol ^{irim}	igneous rim	F093	oxx-ol-chd8@748	lixo-ol-chd8@748	-43.8 (2)	-45.5 (4)	-22.7	-21.2 (1.5)	0.134
PO-8	Ol	mantle	Fo ₉₂	oxx-ol-chd8@749	lixo-ol-chd8@749	-10.0 (2)	-11.4 (5)	-6.2	-8.8 (1.4)	0.181
PO-8	Ol	mantle	Fo ₉₂	oxx-ol-chd8@750	lixo-ol-chd8@750	-18.7 (2)	-19.5 (5)	-9.8	-9.0 (1.5)	0.176
PO-8	Ol	mantle	Fo ₉₅	oxx-ol-chd8@768	lixo-ol-chd8@768	-21.7 (3)	-23.6 (4)	-12.3	-5.8 (1.4)	0.217
PO-8	Ol	mantle	Fo ₉₈	oxx-ol-chd8@769	lixo-ol-chd8@769	-17.2 (2)	-17.9 (5)	-9.0	-5.6 (1.7)	0.143
PO-8	Ol	mantle	Fo ₉₅	oxx-ol-chd8@770	lixo-ol-chd8@770	-30.9 (3)	-33.0 (6)	-16.9	4.9 (0.88)	0.364
PO-8	Ol	mantle	Fo ₉₅	oxx-ol-chd8@771	lixo-ol-chd8@771	-44.7 (2)	-46.0 (4)	-22.8	-10.4 (1.8)	0.0942
PO-8	Ol	mantle	Fo ₉₃	oxx-ol-chd8@772	lixo-ol-chd8@772	-7.2 (2)	-11.5 (5)	-7.8	-9.5 (1.5)	0.144
PO-8	Ol ^{irim}	igneous rim	Fo ₈₉	oxx-ol-chd8@773	lixo-ol-chd8@773	-43.4 (2)	-46.1 (5)	-23.5	-7.5 (1.8)	0.106
PO-8	Olirim	igneous rim	Fo ₈₉	oxx-ol-chd8@774	lixo-ol-chd8@774	-43.0 (2)	-45.5 (5)	-23.1	-3.9 (1.3)	0.2
PO-8	Ol	mantle	F097	oxx-ol-chd8@778	lixo-ol-chd8@778	-17.7 (2)	-19.6 (4)	-10.4	-9.5 (1.0)	0.277
PO-8	Ol ^{irim}	igneous rim	Fo ₈₉	oxx-ol-chd8@779	lixo-ol-chd8@779	-8.7 (2)	-11.5 (5)	-7.0	-13.1 (1.2)	0.316
PO-8	Ol ^{irim}	igneous rim	Fo ₈₉	oxx-ol-chd8@780	lixo-ol-chd8@780	-3.8 (2)	-6.9 (5)	-4.9	-11.7 (1.0)	0.576
PO-8	Ol	mantle	Fo ₉₃	oxx-ol-chd8@781	lixo-ol-chd8@781	-45.7 (2)	-45.9 (4)	-22.1	-9.4 (1.2)	0.243
PO-8	Ol	core	Fo ₈₁	oxx-ol-chd8@801	lixo-ol-chd8@801	-11.3 (2)	-13.2 (5)	-7.3	-6.1 (1.7)	0.109
PO-8	Ol	mantle	Fo ₉₂	oxx-ol-chd8@802	lixo-ol-chd8@802	-13.1 (2)	-14.8 (4)	-8.0	-13.1 (1.6)	0.147
PO-8	Ol	mantle	Fo ₉₆	oxx-ol-chd8@803	lixo-ol-chd8@803	-35.8 (2)	-37.5 (5)	-18.9	-7.8 (1.6)	0.155
PO-8	Ol	mantle	Fo ₉₆	oxx-ol-chd8@804	lixo-ol-chd8@804	-32.6 (2)	-35.4 (5)	-18.4	-11.4 (1.3)	0.215
PO-8	Ol	mantle	Fo ₉₆	oxx-ol-chd8@805	lixo-ol-chd8@805	-8.2 (2)	-12.2 (4)	-7.9	-5.8 (1.7)	0.134
PO-8	Ol	mantle	Fo ₉₆	oxx-ol-chd8@806	lixo-ol-chd8@806	-12.2 (2)	-15.2 (4)	-8.9	-2.7 (1.6)	0.133
PO-8	Ol	mantle	Fo ₉₁	oxx-ol-chd8@822	lixo-ol-chd8@822	-46.5 (2)	-47.9 (5)	-23.7	-7.5 (1.4)	0.164
PO-8	Ol	mantle	F094	oxx-ol-chd8@823	lixo-ol-chd8@823	-36.8 (2)	-38.8 (4)	-19.7	-6.9 (1.7)	0.145
PO-8	Ol	mantle	Fo ₈₅	oxx-ol-chd8@824	lixo-ol-chd8@824	-43.1 (2)	-44.5 (4)	-22.1	-3.4 (1.8)	0.137
PO-8	Ol	mantle	Fo ₉₄	oxx-ol-chd8@828	lixo-ol-chd8@828	-36.1 (2)	-35.8 (5)	-17.0	-13.4 (1.2)	0.217
PO-8	Ol	mantle	Fo ₉₂	oxx-ol-chd8@829	lixo-ol-chd8@829	-37.4(2)	-37.1 (4)	-17.7	-8.6 (1.2)	0.304
PO-8	Ol ^{irim}	igneous rim	Fo ₈₂	oxx-ol-chd8@830	lixo-ol-chd8@830	-7.9 (2)	-10.9 (4)	-6.8	-11.1 (0.86)	0.672
PO-8	Olirim	igneous rim	Fo ₈₇	oxx-ol-chd8@831	lixo-ol-chd8@831	-43.2 (2)	-45.3 (4)	-22.8	-9.4 (1.4)	0.306
PO-8	Olirim	igneous rim	Fo ₈₆	oxx-ol-chd8@833	lixo-ol-chd8@833	-11.7 (3)	-13.9 (4)	-7.8	-14.7 (1.9)	0.0869
PO-8	Ol ^{irim}	igneous rim	Fo ₈₂	oxx-ol-chd8@834	lixo-ol-chd8@834	-42.9 (2)	-44.7 (4)	-22.4	-23.1 (1.7)	0.12
PO-8	Ol	mantle	Fo ₉₁	oxx-ol-chd8@849	lixo-ol-chd8@849	-8.6 (2)	-12.4 (5)	-7.9	-11.0 (1.4)	0.193
PO-8	Ol	mantle	F094	oxx-ol-chd8@850	lixo-ol-chd8@850	-27.4(2)	-29.8 (4)	-15.6	-2.4 (1.1)	0.306
PO-8	Ol	mantle	Fo ₉₄	oxx-ol-chd8@851	lixo-ol-chd8@851	-49.1 (2)	-49.0 (4)	-23.5	-14.5 (1.8)	0.106
PO-8	Ol	mantle	Fo ₉₂	oxx-ol-chd8@852	lixo-ol-chd8@852	-38.6 (3)	-40.3 (5)	-20.2	-6.8 (1.2)	0.233
PO-8	Ol	mantle	Fo ₉₂	oxx-ol-chd8@853	lixo-ol-chd8@853	-21.6(2)	-23.7 (4)	-12.5	-10.3 (1.4)	0.197
PO-8	Ol	mantle	Fo ₉₁	oxx-ol-chd8@854	lixo-ol-chd8@854	-45.2 (2)	-47.1 (4)	-23.6	-10.9 (1.5)	0.154
PO-8	Ol	core	Fo ₈₉	oxx-ol-chd8@858	lixo-ol-chd8@858	-17.0(2)	-20.4 (4)	-11.6	-6.3 (1.4)	0.16
PO-8	Ol	core	Fo ₉₀	oxx-ol-chd8@859	lixo-ol-chd8@859	-23.9 (2)	-26.0 (5)	-13.6	-8.0 (1.1)	0.259
PO-8	Ol	mantle	Fo ₉₅	oxx-ol-chd8@860	lixo-ol-chd8@860	-45.4 (2)	-47.2 (4)	-23.6	-3.9 (1.4)	0.201
PO-8	Ol	mantle	Fo ₉₁	oxx-ol-chd8@861	lixo-ol-chd8@861	-45.1 (2)	-46.5 (4)	-23.0	-5.9 (2.2)	0.0852

Table S4: Continued

	phase	occurrence	compo*	O-analysis ID	Li-analysis ID	δ^{18} O	$\delta^{17}O$	$\Delta^{17}O$	δ^7 Li	$[Li]^{\mu g \cdot g^{-1}}$
PO-8	Ol	mantle	Fo ₉₃	oxx-ol-chd8@862	lixo-ol-chd8@862	-37.6 (2)	-39.0 (4)	-19.4	-3.1 (2.9)	0.044
PO-8	Ol	mantle	Fo ₉₂	oxx-ol-chd8@863	lixo-ol-chd8@863	-38.2 (2)	-40.4 (3)	-20.5	-6.5 (0.90)	0.355
PO-8	LPxirim	igneous rim	$En_{93}Wo_5$	oxy-opx-chd8@9553	liso-opx-chd8@9262	-3.1(1)	-7.1 (3)	-5.4	-2.9 (3.1)	0.591
PO-8	LPxirim	igneous rim	$En_{92}Wo_6$	oxy-opx-chd8@9554	liso-opx-chd8@9263	-0.4(1)	-5.7 (3)	-5.5	0.9(2.1)	1.29
PO-8	HPx ^{irim}	igneous rim	$En_{64}Wo_{35}$	oxy-cpx-chd8@9557	liso-cpx-chd8@9265	-5.5 (1)	-7.4 (3)	-4.5	-17.7 (5.2)	0.113
PO-8	HPx ^{irim}	igneous rim	$En_{66}Wo_{33}$	oxy-cpx-chd8@9558	liso-cpx-chd8@9266	-4.8 (1)	-7.9 (3)	-5.3	-10.1 (3.3)	0.228
PO-8	HPxirim	igneous rim	$En_{57}Wo_{42}$	oxy-cpx-chd8@9559	liso-cpx-chd8@9267	-5.6(1)	-8.6 (3)	-5.7	-2.4 (3.0)	0.214
PO-8	HPxirim	igneous rim	$En_{61}Wo_{38}$	oxy-cpx-chd8@9560	liso-cpx-chd8@9268	-5.7 (1)	-8.7 (3)	-5.7	-4.1 (2.3)	0.258
PO-8	Pl	mantle	$An_{82}Ab_{18}$	oxy-pl-chd8@9549	liso-pl-chd8@9237	4.3(1)	-0.7 (3)	-2.9	-10.1 (4.7)	0.0512
PO-8	Pl	mantle	$An_{83}Ab_{17}$	oxy-pl-chd8@9550	liso-pl-chd8@9238	1.1(1)	-2.8 (3)	-3.3	2.5 (3.8)	0.0591
PO-8	Pl	mantle	$An_{81}Ab_{19}$	oxy-pl-chd8@9551	liso-pl-chd8@9240	4.0(2)	-1.6 (3)	-3.6	-4.6 (2.2)	0.122
PO-3n	Ol	main	Fo ₉₃	oxy-ol-chd3n@9702	liso-ol-chd3n@9062	-8.5 (2)	-11.3 (3)	-6.9	5.2 (2.4)	0.214
PO-3n	Ol	main	Fo ₉₃	oxy-ol-chd3n@9704	liso-ol-chd3n@9096	-23.6 (2)	-25.5 (4)	-13.2	5.5 (3.3)	0.0488
PO-3n	Ol	main	Fo ₉₅	oxy-ol-chd3n@9707	liso-ol-chd3n@9095	-18.7 (2)	-20.7 (3)	-11.0	35.5 (1.9)	0.243
PO-3n	Ol	main	Fo ₇₇	oxy-ol-chd3n@9709	liso-ol-chd3n@9093	-24.6 (2)	-27.1 (3)	-14.3	3.6 (2.3)	0.0984
PO-3n	Ol	main	Fo ₉₄	oxy-ol-chd3n@9710	liso-ol-chd3n@9097	-14.2 (2)	-16.7 (3)	-9.3	4.3 (2.0)	0.119

Abbreviation compo* denotes major-element composition.

Ol, LPx, and HPx are olivine, low-Ca pyroxene, and high-Ca pyroxene at the main chondrule (including mantle or core regions).

 $\overrightarrow{Ol^{irim}}$, $\overrightarrow{LPx^{irim}}$, and $\overrightarrow{HPx^{irim}}$ are olivine, low-Ca pyroxene, and high-Ca pyroxene at igneous rim.

Pl is plagioclase and Ol^{FeO} is olivine with chemical composition $Fo_{\leq 65}$.

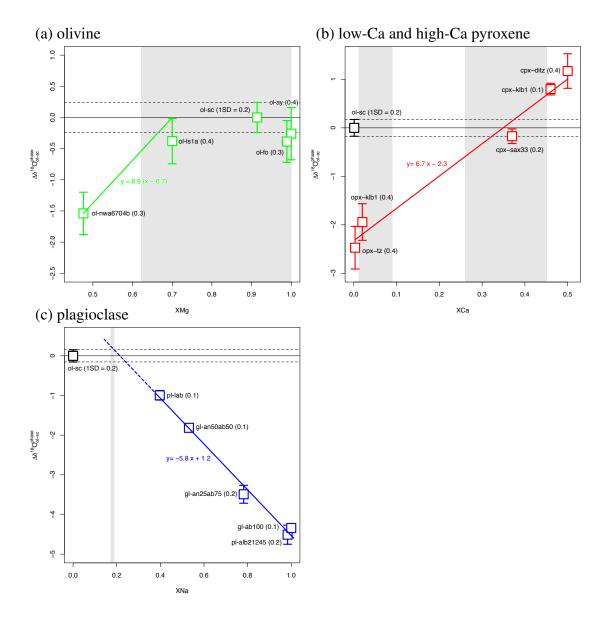


Figure S1: Matrix effects of (a) olivine, (b) low-Ca and high-Ca pyroxene, and (c) plagioclase in O-isotope analyses. Error bars and values in parentheses show 1SD deviations. Shaded regions indicate the compositional ranges of phases analyzed in this study.

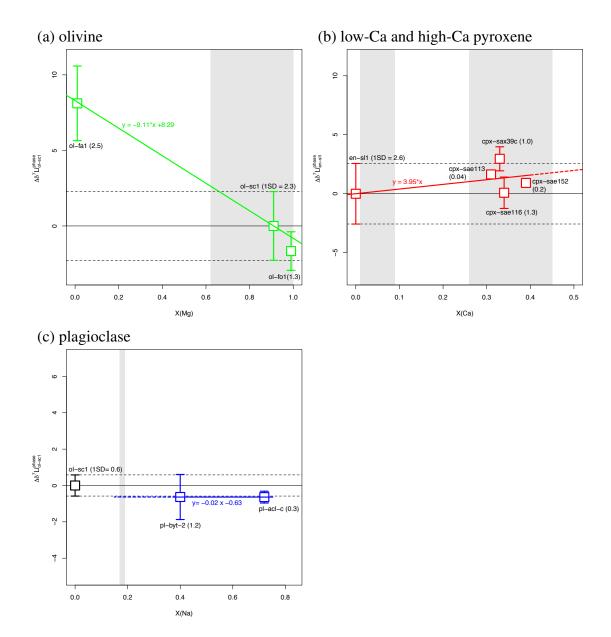


Figure S2: Matrix effects of (a) olivine, (b) low-Ca and high-Ca pyroxene, and (c) plagioclase in Li-isotope analyses. Error bars and values in parentheses show 1SD deviations. Shaded regions indicate the compositional ranges of phases in unknown samples analyzed by this study.

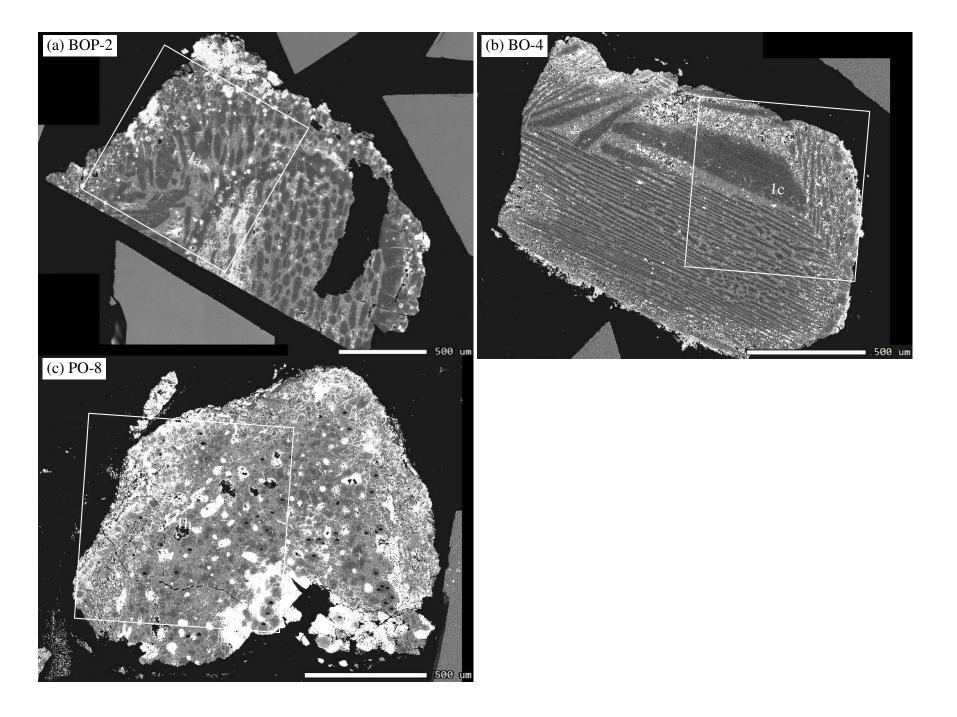


Figure S3: Backscattered electron images of (a) BOP-2, (b) BO-4, and (c) PO-8.

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