

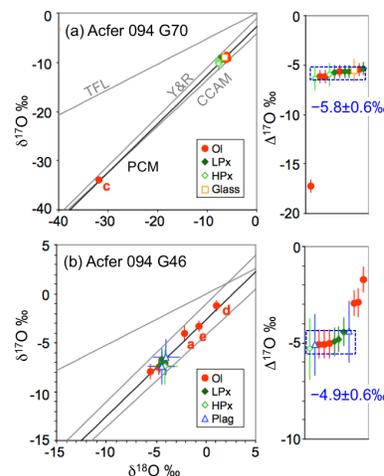
**INTERNAL HOMOGENEITY OF OXYGEN ISOTOPE RATIOS IN CHONDRULES.** N. T. Kita<sup>1</sup>, T. J. Tenner<sup>2</sup>, T. Ushikubo<sup>3</sup>, D. Nakashima<sup>4</sup>, N. G. Rudraswami<sup>5</sup>, M. K. Weisberg<sup>6,7</sup>, C. Defouilloy<sup>1</sup>, M. Kimura<sup>8</sup>, H. Nagahara<sup>9</sup>, and A. Bischoff<sup>10</sup>. <sup>1</sup>WiscSIMS, University of Wisconsin-Madison, Madison, WI 53706. (noriko@geology.wisc.edu). <sup>2</sup>Los Alamos National Laboratory, Los Alamos, NM 87545. <sup>3</sup>Kochi Institute for Core Sample Research, JAMSTEC, Kochi 783-8502, Japan. <sup>4</sup>Tohoku University, Miyagi 980-8578, Japan. <sup>5</sup>National Institute of Oceanography, Dona Paula, Goa 403004, India. <sup>6</sup>Kingsborough College and Graduate Center, CUNY, Brooklyn, NY 11235. <sup>7</sup>American Museum Natural History, New York, NY 10024. <sup>8</sup>Ibaraki University, Mito 310-8512, Japan. <sup>9</sup>University of Tokyo, Tokyo 113-0033, Japan, <sup>10</sup>Institut für Planetologie, WWU Münster, 48149 Münster, Germany.

**Introduction:** Chondrules in primitive chondrites show a range of  $\delta^{17,18}\text{O}$  values mostly from  $-15\text{‰}$  to  $+5\text{‰}$  [e.g., 1], which is significantly depleted in  $^{16}\text{O}$  compared to CAIs ( $-50\text{‰}$  [2]) and the Sun ( $-60\text{‰}$  [3]). Since Clayton et al. [4] reported higher  $\delta^{17,18}\text{O}$  values in chondrules with barred olivine textures (completely melted) than porphyritic ones (partially melted), it is widely accepted that  $^{16}\text{O}$ -rich chondrule precursors exchanged oxygen isotopes with an  $^{16}\text{O}$ -poor nebula gas during melting. By using secondary ion mass spectrometry (SIMS), [5] reported systematically higher  $\delta^{17,18}\text{O}$  values in pyroxene than olivine in individual chondrules, suggesting chondrules formed by interactions between Mg-rich olivine and SiO gas in the nebula. Experiments of oxygen isotope exchange rates between silicate melts and  $\text{H}_2\text{O}$  gas [6-7] suggest oxygen isotope exchange between chondrule melt and nebula gas could be incomplete and resulting chondrules may have internally heterogeneous isotope ratios, as indicated by [5].

At WiscSIMS, we have conducted O-isotope analyses of more than 400 chondrules from 10 different primitive chondrites using an IMS 1280 [8-18]. Within most chondrules (except for CH chondrites with small chondrules  $\leq 30\mu\text{m}$  [18]), multiple high-precision spot data ( $n=4-10$ ) were obtained, in order to test internal heterogeneity of O-isotopes. Here we review this dataset, demonstrating that most ( $>90\%$ ) chondrules have internally homogeneous O-isotope ratios, contrary to conventional wisdom.

**Chondrule dataset:** We used data from 330 chondrules in 9 different chondrite groups in which at least 4 SIMS spot analyses per chondrule were obtained; CO, CV, CR, H3, LL3, R3, EH3, and two ungrouped carbonaceous chondrites Acfer 094 and Y-82094 [8-17]. Most data are from olivine and low-Ca pyroxene phenocrysts analyzed with  $15\mu\text{m}$  spots and with high precision ( $\leq 0.5\text{‰}$  in 2SD for  $\delta^{17,18}\text{O}$ ) using multiple Faraday cups. Data from high Ca-pyroxene and plagioclase were also obtained for some chondrules. In Acfer 094 and LL3, small spots ( $3-5\mu\text{m}$ ) with moderate precision ( $\sim 1\text{‰}$  in 2SD for  $\delta^{17,18}\text{O}$ ) were used for small ( $<10\mu\text{m}$ ) olivine and pyroxene phenocrysts and the mesostasis phases, including glass [8, 11].

**Internal Homogeneity:** Fig. 1 shows examples of SIMS data for two chondrules G70 and G46 from Acfer 094 [11]. They have relict olivine grains with variable O-isotope ratios ( $^{16}\text{O}$ -rich in G70 and  $^{16}\text{O}$ -poor in G46). Excluding relict olivine, data from multiple phases (olivine, low- and high-Ca pyroxene, plagioclase or glass) are homogeneous in  $\Delta^{17}\text{O}$  ( $\delta^{17}\text{O} - 0.52 \times \delta^{18}\text{O}$ ), with variability similar to the analytical reproducibility ( $0.5-1\text{‰}$  in 2SD). Thus, host chondrule O-isotope ratios are determined precisely as an average of these multiple analyses (excluding relict olivine).

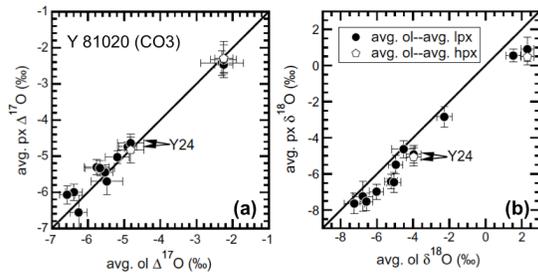


**Fig. 1.** O-isotope in chondrules (a) G70 and (b) G46 from Acfer 094 [11]. Terrestrial fractionation line (TFL) and three slope  $\sim 1$  lines (CCAM, Y&R, and PCM [2, 11 19]) are shown. The average  $\Delta^{17}\text{O}$  (and 2SD) from homogeneous data are shown.

Among 40 chondrules investigated in Acfer 094, 90% of them have multiple data with indistinguishable  $\Delta^{17}\text{O}$  values (2SD  $< 1\text{‰}$ ) [11]. Among other chondrite groups 80-90% of their chondrules are also homogeneous. The frequency of chondrules with relict olivine grains differs among chondrite groups:  $\sim 50\%$  in Acfer 094, CO3, and Y-82094 [11-12, 17], and uncommon ( $< 20\%$ ) in other chondrites.

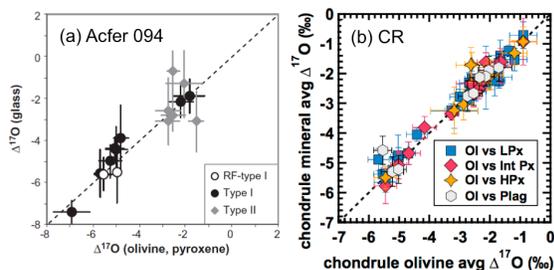
**Olivine vs. Pyroxene:** O-isotope data of coexisting olivine and low-Ca pyroxene can be compared among 198 chondrules in the dataset. For example, in CO3 chondrite chondrules (Fig. 2a), averaged  $\Delta^{17}\text{O}$  values of coexisting olivine and low-Ca pyroxene are in agreement, plotting on a 1:1 line. Similar agreements between  $\Delta^{17}\text{O}$  values of coexisting olivine and pyroxene are observed for chondrules in other chondrite

groups. In Fig. 2b,  $\delta^{18}\text{O}$  values plot slightly off the 1:1 line towards higher  $\delta^{18}\text{O}$  in olivine relative to pyroxene, suggesting small natural mass fractionation effects ( $\leq 1\%$ ), possibly due to evaporation and recondensation of oxides during chondrule melting [20]. These data are very different from those of [5], who claimed systematically lower  $\delta^{17,18}\text{O}$  values along a slope-1 line (thus also lower  $\Delta^{17}\text{O}$ ) in olivine versus pyroxene.



**Fig. 2.** Comparison between O-isotope ratios of olivine and pyroxene from chondrules in Y-81020 (CO3) [12].

**Phenocrysts vs. Mesostasis:**  $\Delta^{17}\text{O}$  values in Acfer 094 chondrule glass (Fig. 3a) have a 1:1 relationship to coexisting olivine and pyroxene data [11]. Similarly,  $\Delta^{17}\text{O}$  values of mesostasis high-Ca pyroxene and plagioclase in CR chondrules are consistent with those of coexisting olivine and low-Ca pyroxene [13] (Fig. 3b). Similar results are also obtained from Y-82094 [17]. Thus, O-isotope ratios of chondrule mesostasis are in good agreement with those of coexisting olivine and pyroxene, suggesting O-isotope ratios in chondrule melts did not change during subsequent cooling.



**Fig. 3.** The  $\Delta^{17}\text{O}$  values in chondrule mesostasis in the least metamorphosed chondrites. (a) glass in Acfer 094 compared to olivine and pyroxene phenocrysts [11]. (b) Comparison between olivine and other minerals in chondrules from CR [13].

In contrast to the data mentioned above,  $\Delta^{17}\text{O}$  values of chondrule glass in LL3 chondrites and those of plagioclase in Allende chondrites are significantly higher than those of coexisting olivine and pyroxene [8, 10]. Acfer 094 and CR chondrites are among the least aqueously altered and thermally metamorphosed, and likely retained primary O-isotope signatures. LL3

and Allende chondrites experienced low degree aqueous alteration and thermal metamorphism, in which primary O-isotope ratios of glass and plagioclase were likely disturbed because oxygen diffusion rates in these phases are much faster than olivine and pyroxene [21].

**Formation Environments:** Chondrules likely formed in the protoplanetary disk with dust to gas ratios at least  $100\times$  solar [e.g., 13, 20]. If chondrules formed in an open system with significant evaporation and re-condensation of oxide from dust [20], then the source of oxygen in the ambient gas would be dominated by that from solid precursors [8, 11]. If true, then both the melt and ambient gas would have isotope ratios very similar to those of the averaged precursor dusts. Unmelted relict olivine grains in porphyritic chondrules would have preserved their distinct O-isotope signatures due to the slow diffusion rate of oxygen in olivine [21], and based on their signatures, some relict olivines originated from  $^{16}\text{O}$ -rich refractory precursors [1]. However, most relict olivines have likely originated from previous generations of chondrules [11-13] that already had O-isotope ratios much closer in value to the newly formed melt.

The average  $\Delta^{17}\text{O}$  values of chondrules in each chondrite group estimated from SIMS generally show restricted ranges that are specific to each group, such as  $-5\%$  and  $-2\%$  in Acfer 094 and CO, and  $+1\%$  in LL, H, and R. These values may represent the average O-isotope ratios of the local areas of the disk, which further constrain the evolution of isotope reservoirs in the protoplanetary disk of our Solar System.

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