Chondritic meteorites are composed of primitive components formed during the evolution of the Solar protoplanetary disk. The oldest of these components formed by condensation, yet little is known about their formation mechanism because of secondary heating processes that erased their primordial signature. Amoeboïd Olivine Aggregates (AOAs) have never been melted and underwent minimal thermal annealing, implying they might have retained the conditions under which they condensed. We performed a multiisotope (O, Si, Mg) characterization of AOAs to constrain the conditions under which they condensed and the information they bear on the structure and evolution of the Solar protoplanetary disk. High-precision silicon isotopic measurements of 7 AOAs from weakly metamorphosed carbonaceous chondrites show large, mass-dependent, light Si isotope enrichments (−9% < δ63Si < −1%). Based on physical modeling of condensation within the protoplanetary disk, we attribute these isotopic compositions to the rapid condensation of AOAs over timescales of days to weeks. The same AOAs show slightly positive δ26Mg that suggest that Mg isotopic homogenization occurred during thermal annealing without affecting Si isotopes. Such short condensation times for AOAs are inconsistent with disk transport timescales, indicating that AOAs, and likely other high-temperature condensates, formed during brief localized high-temperature events.

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Significance

Combined O, Si, Mg isotopic measurements of amoeboïd olivine aggregates allow the individual condensation timescales of a typical set of the first Solar System solids to be estimated. Our results indicate formation over days or weeks, much faster than could be inferred from the secular evolution of the Solar protoplanetary disk. The oldest solids of the Solar System thus bear witness to a turbulent disk with strong thermal heterogeneities.

Author contributions: Y.M. and J.V. designed the study and performed the isotopic measurements; Y.M., J.V., E.J., M.P., and M.C. discussed the ion probe data; E.J. devised the astrophysical model; Y.M., J.V., E.J., and M.C. wrote the paper.

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implications on the structure and evolution of the Solar protoplanetary disk.

**Material and Methods**

We surveyed all AOAs in a section each of Kaba (thin section N4075 from the Natural History Museum, Vienna, Austria), Northwest Africa (NWA) 5958 (thick section NWA 5958-1 from the Museum national d'Histoire naturelle, Paris, France) and Miller Range (MIL) 07342 (thick section MIL 07342,9 from the NASA Antarctic Search for Meteorites program). Kaba is an oxidized Bali-like CV chondrite (28, 29). Although opaque assemblages are altered and evidence of incipient metasomatism appears on chondrule mesostasis, olivine compositions in type 1 chondrules (e.g., Figs. S4-S9) show no diffusional exchange; the chondrite is deemed of type 3.0–3.2 by the Antarctic Meteorite Petrographic Description database. NWA 5958 is a C2-ung CM-like chondrite, yet chondrule mesostases have undergone extensive aqueous alteration, which did not affect olivine (30, 31).

We analyzed the silicon isotopic compositions of olivines by secondary ion mass spectrometry (SIMS) using the ultrahigh-resolution mass spectrometer CAMECA SX-100 (at CAMPARIS, Sorbonne Université, Paris, France). A 20-nA focused beam accelerated to 15-kV potential difference was used for spot analyses of olivine with 20-s analysis times. The PAP software was used for matrix corrections.

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Scanning electron microscope observations of AOAs were performed at CRRG-CNRS (Nancy, France) using a JEOL JSM-6510 with three-nA primary beam at 15 kV. Quantitative compositional analyses of olivine in AOAs were performed using a CAMECA SX-100 electron microprobe (at CAMPARIS, Sorbonne Université, Paris, France). A 20-nA focused beam accelerated to 15-kV potential difference was used for spot analyses of olivine with 20-s analysis times. The PAP software was used for matrix corrections.

**Results**

AOAs in NWA 5958 and MIL 07342 have compact textures characterized by continuous olivine (Fig. 1 and SI Appendix, Figs. S1 and S2) (33). Ca–Al-rich vermicular patches are altered, although diopside persists, in AOAs in NWA 5958. AOAs in Kaba show smaller olivine zones, diopside, and Ca–Al-rich areas (SI Appendix, Fig. S3). Olivine in all AOAs investigated herein are Mg-rich with Mg# in the range 98.9 to 99.9 (SI Appendix, Table S1). AOAs olivine grains have O isotopic compositions with $\delta^{18}$O values ranging from −48.6 to −39.8‰, $\delta^{17}$O values ranging from −47.7 to −41.5‰, and $\Delta^{17}$O values ranging from −23.9 to −19.2‰ (SI Appendix, Figs. S4 and Table S2). The silicon isotopic compositions of OAAs olivines show large, mass-dependent isotopic variations with $\delta^{30}$Si values ranging from 5.1 to 1.0‰ and $\delta^{32}$Si values ranging from 9.7 to −2.2‰ (Figs. 1 and 2 and SI Appendix, Table S3). We did not observe any correlation between the Si compositions and the analytical location within the AOAs (Fig. 1 and SI Appendix, Figs. S1 and S2). The magnesium isotopic compositions of olivines in AOAs show slightly positive $\delta^{29}$Mg values ranging from 0.09 to 1.81‰ (Fig. 3 and SI Appendix, Table S4). We did not observe any correlation between the average Si and Mg isotopic compositions of the AOAs, nor with their elemental compositions.

**Duration and Contexts of AOA Condensation.** The mass-dependent character of the Si isotopic variations precludes their explanation by nucleosynthetic anomalies in the AOA-forming region. Provided bulk carbonaceous chondrites are a suitable proxy for the composition of the reservoir from which AOAs formed, the light isotope enrichment is also at variance with the positive 2‰/$\alpha$ fractionation expected from olivine-gas equilibrium (34) and inferred by ref. 15 to account for the Si isotopic variations of bulk chondrite. Partial evaporation would likewise produce heavy compositions in olivine, unless it occurred subordinately

![Fig. 1. A representative AOA and the Si isotopic compositions of its olivines. (A) Back-scattered electron image of AOA N1-8 from the CM-related chondrite NWA 5958 (30, 31). SIMS measurements are indicated by colored ellipses and the Si isotopic compositions associated with each analysis point. The color of the ellipses indicates the variability of the Si isotopic compositions by the range of the $\delta^{30}$Si value: blue, $-3\% < \delta^{30}$Si $< 0\%$, orange, $-6\% < \delta^{30}$Si $< -3\%$, and red, $\delta^{30}$Si $< -6\%$. (B) $\delta^{30}$Si – $\delta^{32}$Si diagram showing the mass-dependent variation observed in N1-8; MDFL, mass-dependent fractionation line.](https://www.pnas.org/doi/10.1073/pnas.1912479116)

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on isotopically lighter objects. In contrast, light isotopic enrichment would be expected from condensation, as the light isotopomers of gaseous SiO would collide with the growing solids at greater rates proportional to their thermal velocity (e.g., ref. 20)

$$v_{T, SiO} = \sqrt{\frac{8 k T}{\pi m_{SiO}}}$$

where $k$ is the Boltzmann constant, $T$ is the temperature, and $m_{SiO}$ is molecular mass. We note that the greatest observed Si isotopic fractionations, around $-5\% / \mu$ (Fig. 2 and SI Appendix, Table S3), are not far from the maximum kinetic fractionations of $-11\% / \mu$, assuming that the condensation coefficient $\gamma_{SiO}$ has no mass dependence (SI Appendix, Fig. S5). This indicates back-evaporation has had little opportunity to curtail the isotopic fractionation of the condensate. This is possible if the reservoir temperature evolved through the olivine condensation interval over a time shorter than the “SiO condensation timescale,” that is, the theoretical (e-folding) time of SiO condensation (on the available solids) in the absence of evaporation, given by (20) (SI Appendix):

$$t_{cond} = \frac{1}{n_p r a^2 \gamma_{SiO} v_{T, SiO}}$$

where $n_p$ is the number density of condensate particles, assumed to be identical spheres of density $\rho_s = 3 \times 10^3$ kg/m$^3$ and radius $a$. If indeed the reservoir temperature evolved more rapidly than $t_{cond}$, high supersaturation would develop and make back-evaporation subdominant. In Fig. 4, we show the distribution of $\delta^{30}\text{Si}$ expected for different cooling timescales $\lambda^{-1}$ ($\lambda$ being defined as the decay constant of the saturation density of Mg, related by stoichiometry to SiO; see SI Appendix). It is seen that only $\lambda_{cond} > 1$, that is, $\lambda^{-1}$ values shorter than $t_{cond}$, can reproduce the range of the data. Thus, AOAs must have formed over a timescale of $\max(\lambda_{cond}, \lambda^{-1}) = t_{cond}$.

So what is the magnitude of $t_{cond}$? For a steady $\alpha$ disk of mass accretion rate $M$, using the pressure expression (SI Appendix), we have

$$t_{cond} = \frac{m_{SiO}^{1/2}}{327_{SiO} m_g T^{7/3}} \left( \frac{k \rho_a a M}{\bar{e}_p} \right)^{1/3} \left( \frac{9 \pi}{\sigma} \right)^{2/3}$$

$$= 10^5 s \left( \frac{0.1}{\gamma_{SiO}} \right)^{1/3} \left( \frac{\alpha}{10^{-5}} \right)^{1/3} \left( \frac{M}{10^{-7} M_\odot / a} \right)^{1/3} \times \left( \frac{10^{-3}}{\bar{e}_p} \right)^{1/3} \left( \frac{1400 K}{T} \right)^{7/3},$$

where $\alpha$ is the turbulence parameter (35), $\sigma$ is the Stefan–Boltzmann constant, $m_g = 2.34$ u is the mean nebular gas molecular mass, $\bar{e}_p$ is the solid/gas mass ratio, and $M_\odot$ is the solar mass. Quite insensitively to the values of the disk parameters, our calculations thus reveal a short duration on the order of a day ($\sim 10^5$ s) for the condensation of AOAs, or weeks if smaller values of $\gamma_{SiO}$ apply (e.g., ref. 36 and Fig. 4). This is consistent with the cooling rate lower limit of 0.02 K/h derived by (17) from the quasi-steady absence of low-Ca-pyroxene in AOAs, as well as the 50 K/h cooling rate suggested for the silica-bearing AOA of ref. 18. This may also account for the disequilibrium gas–solid condensation effects described for AOAs in Allan Hills 77037 (24).

Such durations are, however, much shorter than the $10^3$ to $10^5$ y timescales of secular evolution (and cooling) of the disk as a result of progressive accretion into the Sun (13). They are also much shorter than radial transport timescales, which ref. 37 estimated on the order of $10^7$ y (SI Appendix). Even vertical transport toward cooler surface layers, away from the disk mid-plane, would take much longer than the orbital period (38), even if winds are taken into account (SI Appendix). However, the smooth temperature profiles generally invoked in the inner disk are only average; heating would have been dictated by the dissipation of turbulence and likely prone to local heterogeneities, for instance in current sheets in magnetorotational instability-driven turbulence (39–41). Outside the “nominal” condensation front of forsterite, transient temperature excursions may have led to the olivine vaporization and recondensation. Perhaps late mixing with isotopically unfractinated SiO from the surroundings prevented the positive isotopic compositions predicted at the end of closed-system condensation (Fig. 4) but not observed in the data, although we cannot exclude an effect of the finite size of the ion probe beam (SI Appendix). In our model (illustrated schematically in Fig. 5), AOAs would have formed at

![Fig. 2. Silicon 3-isotope plot showing the large range of mass-dependent isotopically light values measured in all analyzed AOAs in NWA 5958 (red), MIL 07342 (black), and Kaba (blue).](https://example.com/fig2)

![Fig. 3. $\delta^{26}\text{Mg}$ values of AOAs analyzed in NWA 5958, MIL 07342, and Kaba.](https://example.com/fig3)
greater heliocentric distances than a mere consideration of the average isotherms would suggest. Those which we now see in chondrites would have eventually escaped the evaporation/recondensation cycle of their matter during outward migration toward less-heated regions, e.g., as a result of turbulent diffusion or disk expansion (13, 42, 43). The “frozen-in” AOAs would then have eventually reached the cold regions of chondrite accretion.

Similar processes would likely hold for higher-temperature condensates, i.e., CAIs, at shorter heliocentric distances. There may thus be a continuum between the condensation processes and subsequent reheating events that led to their remelting and crystallization, with cooling rates on the order of 0.1 to 10 K/h (8)—not to mention chondrule-forming processes as well, with cooling rates estimates spanning 1 to 3,000 K/h (ref. 44 and references therein)—the formation of Wark–Lovering rims (45–47) and/or partial evaporation resulting in heavy isotopic enrichments (19). The isolation of the ultrarefractory component presumably lost from the parent reservoir of numerous group II CAIs (48) may have likewise required similarly short timescales. Indeed, the light isotopic enrichment of (refractory) heavy rare-earth elements in such CAIs (49) allows for only limited recondensation during the partial evaporation of the now lost ultrarefractory complement.

Condensate Agglomeration. It is instructive to compare the condensation timescale calculated above to the grain–grain collision timescale \( t_{\text{coll}} \)—that is, the mean time between 2 collisions for a given grain, given by ref. 50:

\[
\frac{t_{\text{coll}}}{t_{\text{cond}}} = \frac{\gamma_{\text{SiO}} T_{\text{SiO}}}{4 \Delta v_{\text{coll}}} = 20 \left( \frac{\gamma_{\text{SiO}}}{0.1} \right) \left( \frac{T}{1,400 \text{ K}} \right)^{1/2}.
\]

Since \( \Delta v_{\text{coll}} \) is normalized to an upper bound (the critical velocity for agglomeration which might have been greater for higher temperatures (52)), generally occurred after, not during, condensation (Fig. 5). Indeed, unmelten fine-grained CAIs are composed of mineralogically zoned sub–100-μm nodules with more refractory minerals concentrated in their center, consistent with an origin as individual condensates, which encompassed the whole refractory condensation sequence before coagulation (53). Still, because \( t_{\text{coll}} \) should be much shorter than radial or vertical transport timescales (14), agglomeration should have occurred largely between genetically related bodies. This accounts for the generally fairly uniform character of fine-grained CAIs at mesoscale, notwithstanding common macroscale layering (53, 54). Same holds for AOAs. One can envision that the high-temperature events that ultimately led to their final condensation likely first evaporated all but the most refractory (CAI-like) phases of any dust grains present. Olivine recondensation would have then mantled these dust grains; perhaps some olivine also condensed homogeneously (such as the \( 16\text{O} \)-rich ones found in chondrite matrices, refs. 55 and 56). These different olivine-bearing bodies would have then aggregated together. While, as argued above, aggregated bodies might have genetic relationships to one another, they may represent different thermal events, if \( t_{\text{coll}} \) was longer than transport timescales or evaporation/recondensation cycles, and thus possibly variable thermal histories. Thus, individual AOAs may group bodies with similar or various ranges in Si isotopic fractionation. Depending on the recondensation events sampled by individual AOAs, different AOAs may vary in their ranges of \( \delta^{30}\text{Si} \).

Secondary Heating of Amoeboid Olivine Aggregates. Since \( \delta^{30}\text{Mg} \) corresponds to similar isotope mass ratios as \( \delta^{25}\text{Si} \) (in terms of Mg and SiO, respectively), one would expect similar fractionations for the former in AOAs (SI Appendix). Mg isotope fractionations as large as –14‰/°C have been reported in fine-grained CAIs in Allende (57, 58). However, we observe little variation of \( \delta^{25}\text{Mg} \) in the AOAs analyzed herein (only 0.4‰/°C SD), although (59) reported \( \delta^{25}\text{Mg} \) down to –3‰/°C in some porous AOAs in Kaba. The compact textures of our AOAs suggest significant postagglomeration annealing, which may have led to the Mg isotopic homogenization given the much higher diffusivity of Mg relative to Si (60). Indeed, for a Mg diffusion coefficient of \( \sim 10^{-17} \text{ m}^2/\text{s} \) at 1,400 K, isotopic homogenization over lengths of 50 μm requires only an integrated (possibly discontinuous) time of a decade or so (Fig. 5), whereas O and Si homogenization requires durations of 6 orders of magnitude longer. A timescale of a decade would agree with 1) the <60 y of isotopic exchange with \( ^{18}\text{O} \)-poor gas for 2 Levolive AOAs (61) and 2) the effective cooling rate of 0.002 K/h estimated from the closure of Mn and Cr diffusion in AOA olivine (17). Ca from the Ca–Al-rich nodules would have diffused as well into forsterite (17, 31), accounting for its apparent Ca enrichment in compact AOAs (3, 17, 31), which our Si isotopic results no longer allow to explain by a condensation slower than for porous AOAs (17).

The closure of the Mg isotopic system in AOAs would postdate the condensation and aggregation period but would not have been as late as parent body metamorphism, because AOAs retain near-canonical Al–Mg mineral isochrons (e.g., refs. 62 and 63). The Mg isotopic composition of the bulk olivine (to which all olivines would have been homogenized) would have been also closer to the reservoir average than that of Si because Mg would be essentially completely condensed during olivine condensation,
while almost half of the original SiO would still remain (ref. 6 and SI Appendix).

In conclusion, our results suggest that the earliest solids of the Solar System condensed, aggregated, and were processed over short timescales, perhaps on the order of days, in a dynamic and thermally nonuniform disk. Nowhere was the primordial condensation sequence quiescent.

Data Availability. All of the data are available in the SI Appendix (SI Appendix, Tables S1–S4). All raw SIMS data, summary of the data, and output of the model can be accessed here: https://data.mendeley.com/datasets/dmxgbzmkz5/.

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Fig. 5. Schematic representation of AOA formation. Localized heating beyond the nominal olivine condensation front may have evaporated solids, leaving refractory residues. Upon cooling, such CAI-like materials served as seeds for olivine condensation slightly beyond the nominal olivine condensation front. The rapid condensation inferred herein (day to weeks) would have produced large negative mass-dependent silicon isotopic fractions. Agglomeration of AOA occurred over a longer timescale on the order of a year. Subsequent thermal annealing produced the AOAs as we observe them today, homogenizing their Mg isotopic compositions without affecting their silicon isotopic compositions.

Table S1. Amoeboid olivine aggregates and related objects in carbonaceous chondrites: Records of nebular and asteroid processes.

| Object | Description | Reference |
|--------|-------------|-----------|
| CAI    | Calcium-aluminum-rich inclusions | Geochim. Cosmochim. Acta 64, 185–239 (2004) |
| AOAs   | Amoeboid olivine aggregates | Geochim. Cosmochim. Acta 64, 185–239 (2004) |
| EVs    | Evaporated vesicles | Geochim. Cosmochim. Acta 64, 185–239 (2004) |

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