



A unifying model for the accretion of chondrules and matrix

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The so far unique role of our Solar System in the universe regarding its capacity for life raises fundamental questions about its formation history relative to exoplanetary systems. Central in this research is the accretion of asteroids and planets from a gas-rich circumstellar disk and the final distribution of their mass around the Sun. The key building blocks of the planets may be represented by chondrules, the main constituents of chondritic meteorites, which in turn are primitive fragments of planetary bodies. Chondrule formation mechanisms, as well as their subsequent storage and transport in the disk, are still poorly understood, and their origin and evolution can be probed through their link (i.e., complementary or noncomplementary) to fine-grained dust (matrix) that accreted together with chondrules. Here, we investigate the apparent chondrule–matrix complementarity by analyzing major, minor, and trace element compositions of chondrules and matrix in altered and relatively unaltered CV, CM, and CR (Vigarano-type, Mighei-type, and Renazzo-type) chondrites. We show that matrices of the most unaltered CM and CV chondrites are overall CI-like (Ivuna-type) (similar to solar composition) and do not reflect any volatile enrichment or elemental patterns complementary to chondrules, the exception being their Fe/Mg ratios. We propose to unify these contradictory data by invoking a chondrule formation model in which CI-like dust accreted to so-called armored chondrules, which are ubiquitous in many chondrites. Metal rims expelled during chondrule formation, but still attached to their host chondrule, interacted with the accreted matrix, thereby enriching the matrix in siderophile elements and generating an apparent complementarity.

complementarity | chondrules | secondary alteration | LA-ICPMS

The blueprint of our Solar System was mainly attained in the turbulent first million years of its lifetime. In this active period, the young Sun was surrounded by a mostly gaseous protoplanetary disk from which planetary bodies formed through accretion of dust and pebble-like objects such as chondrules (1). Along with a fine-grained matrix of dust, chondrules are the main components of chondrites, the most unaltered fragments of asteroids that present us with a 4.5-billion-year window in time toward the early evolution of the Solar System. Chondrules are submillimeter- to centimeter-sized silicate spherules that formed in the protoplanetary disk by thermal processing of preexisting nebular dust. To date, there is no consensus on how, where, and when chondrules were formed, stored, and transported, even though they dominate the meteorite fabric and are the likely building blocks of planets (1). Several mechanisms have been proposed to explain chondrule formation (2), but these have to conform to an increasing list of constraints posed by their isotope signatures and elemental compositions. To complicate matters, some constraints seem to contradict each other. Chondrules are thought to have formed throughout the lifetime of the protoplanetary disk, and their individual ages show a range of ~ 4 Myr using Pb–Pb dating techniques (3, 4). These spherules, which are located centimeters apart within the same meteorite, have varying ages and nucleosynthetic isotope signatures (i.e., Cr and Mg; refs. 5, 6), suggesting they formed at different times and/or

locations before they were transported to their accretion regions (7). In stark contrast, for the same chondrites, an apparent elemental and isotope bulk complementarity is proposed to exist between the matrix and the chondrules (8). Complementarity is a model that invokes chondrules and matrix within a chondrite group to be genetically related and formed from a single, initially CI-like (i.e., and Ivuna-type chondrite that is compositionally similar to the Sun's photosphere) reservoir. Elements lost during chondrule formation are reabsorbed by CI-like dust, generating elemental ratios which are subchondritic (below the CI ratio) in chondrules and superchondritic (above the CI ratio) in matrix. These complementarities are suggested for Fe/Mg, Si/Mg, Ca/Al, Ca/Ti, and Hf/W ratios (8–11) and require a CI-like bulk composition of the analyzed chondrites. Furthermore, other authors argue that in favor of complementarity, chondrule and matrix separates from CV chondrites (i.e., Vigarano-type carbonaceous chondrites) show mirroring W and Mo isotope signatures relative to the bulk chondritic value (11, 12), and volatility patterns of carbonaceous chondrite bulk and matrix are also interpreted in favor of complementarity (13). These conflicting findings—individual chondrule ages and isotope signatures versus bulk chondrule and matrix isotope and chemical composition—leave the cosmochemical community at an impasse. Without a solution to resolve the apparent chondrule–matrix complementarity with respect to the observed temporal and spatial interval over which chondrules formed, progress cannot be made at frontiers that matter to understand the history of our Solar System. This includes understanding the nature of planetary building blocks

Significance

An impasse exists between chemical and astrophysical models that explore the accretion of the Solar System's building blocks, the chondrites. To resolve this issue means to gain an understanding of the dimensions of mass transport in the early Solar System and, hence, a crucial insight into the volatile inventory of the terrestrial planets. Here, we use element volatility patterns of chondrules and their dust rims to show that these main constituents of chondrites are not complementary to each other and did not form in the same chemical reservoirs. We propose a unifying chondrule and matrix accretion model that necessitates significant mass transport in the protoplanetary disk and an inward flux of volatile-rich CI-like (Ivuna-type carbonaceous chondrite) dust.

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Data deposition: Excel data sheet including raw data of LA-ICPMS analyses performed during this study is provided as [Dataset S1](#).

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and the dimensions of mass transport in the protoplanetary disk. One solution is that this apparent complementarity is the result of secondary alteration processes on the chondrite parent bodies (14). Elemental exchange during fluid mobilization may create complementary patterns, whereas the initial composition of the matrix was CI-like. Alternatively, the observed complementarity in chondrites may simply reflect a generic process of chondrule formation that does not require formation of chondrules and matrix from the same reservoir (7, 15). To distinguish between these different models, we have investigated major, minor, and trace element compositions of chondrules and their fine-grained dust rims from altered and relatively unaltered CV (Leoville CV3.1, Vigarano CV3.1-3.4, Allende CV>3.6), CM (Mighei-type; Maribo CM>2.8, Cold Bokkeveld CM2.2), and CR (Renazzo-type; NWA801 CR2) chondrites using scanning electron microscopy (SEM) and laser ablation inductively coupled plasma mass spectrometry (LA-ICPMS). Considering most chondrites are accretionary breccias and contain clasts with various degrees of alteration, we investigate the matrix rims surrounding chondrules, rather than the bulk matrix. If complementarity exists, individual dust rims should be complementary at least to their host chondrule. As such, we provide first-order, model-independent observations between individual chondrules and their accreted matrix. Also in contrast to previous compositional studies of chondritic matrix (13), we include some of the least-altered carbonaceous chondrites and apply the same technique to chondrules as well as matrix. We will discuss the effect of aqueous alteration and thermal metamorphism on the composition of chondrules and surrounding matrix and, finally, a mechanism to unify the apparent elemental complementarity of chondrules and matrix (8) with observed isotope heterogeneities and ages of individual chondrules (3–6). We invoke a unifying model for the accretion of chondrules and matrix, in which their relationship to one another is defined in a 3-component system.

The Complementarity Paradox

Followers of the complementarity model put forward the Fe/Mg and Si/Mg compositions of chondrules and matrix as major cornerstones of the debate. Chondrules typically have subchondritic Fe/Mg ratios, whereas matrix has superchondritic ratios (8). Arguments against these data reflecting complementarity are that 1) the Fe/Mg ratios of chondritic components are usually measured by defocused beam analyses, supposedly biased toward higher Fe/Mg ratios for the matrix, and 2) that these ratios may reflect remobilization through aqueous alteration (14). Here, we have determined the major element composition of CV, CM, and CR chondrules and their dust rims (for a detailed description of their petrology, see *SI Appendix, Supporting Information Text and Figs. S1–S6*). We show that the most unaltered carbonaceous chondrites (Maribo and Leoville) have matrices with superchondritic Fe/Mg ratios, where CM chondrite matrix is closer to the CI composition than CV chondrites (Fig. 1A). More-altered chondrites, having experienced either metamorphism (Allende) or aqueous alteration (Cold Bokkeveld), have increasingly lower Fe/Mg ratios that move toward the CI ratio line and even become subchondritic. The chondrules of all chondrites studied here have subchondritic ratios and CV chondrules have lower Fe/Mg ratios than CM chondrules (Fig. 1B). These trends are independently confirmed by LA-ICPMS data in this study, wet analyses and Instrumental Neutron Activation Analysis for the same chondrites (*SI Appendix, Fig. S7*), bypassing potential analytical artifacts from electron microprobe analyses (14). Even though Fe/Mg ratios of chondrules and matrix from several carbonaceous chondrite groups appear to be in agreement with a complementarity scenario, we emphasize that chondrites are regarded here as 2-component systems of chondrules and matrix, which, as we show below, is clearly not the case. The Si/Mg ratios observed for chondrites in this study show

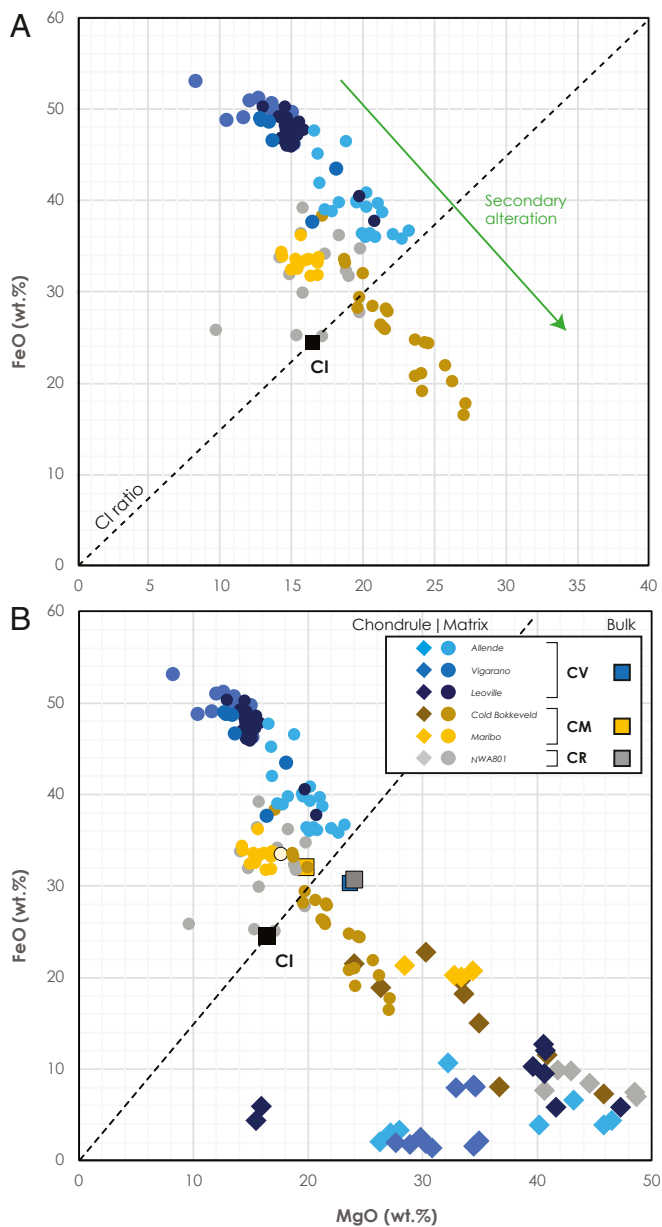


Fig. 1. Major element plots from SEM/EDS elemental maps of CV, CM, and CR chondrules and surrounding dust rims (*SI Appendix, Figs. S1–S6*). (A) FeO versus MgO wt.% of dust rims, reflecting a decrease in Fe/Mg ratio with secondary alteration, and a primary superchondritic composition for the matrix. (B) Same plot as A, but with chondrule and bulk compositions added, which are all subchondritic. Individual data points reflect regions of interest where LA-ICPMS spots were analyzed (*SI Appendix, Dataset S1*). For each sample, this amounts to 3 to 4 chondrules, with 2 to 4 spots per chondrule core and 2 to 4 spots per surrounding dust rim.

that unaltered CV and CM matrices are slightly superchondritic, whereas the chondrules are spread around the CI ratio line and are not definitively subchondritic (*SI Appendix, Fig. S7C*). The CR chondrite displays a more recognizable complementarity (*SI Appendix, Fig. S7D*). One of the main arguments against complementarity is the overall CI-like composition of the Paris matrix (16), Paris being one of the least-altered CM chondrites (CM2.7–2.9). Although the matrix is not perfectly CI-like for all elements (e.g., the Fe/Mg ratio of Paris is also superchondritic), it reflects an absence of volatile addition to a CI-like matrix, as envisioned for the complementarity scenario. During chondrule formation,

nonrefractory main component elements with condensation temperatures $<1,350$ K are lost increasingly according to their relative volatility, resulting in chondrule element-volatility patterns with $>CI$ refractory elements (i.e., the refractory plateau) and $<CI$ moderately volatile elements (Fig. 2A). According to complementarity, the lost volatiles should be reabsorbed by an initially CI-like matrix, resulting in a mirrored volatility pattern relative to the bulk (i.e., chondrule and matrix, without CAIs [calcium-aluminum inclusions], metal etc.; ref. 13). However, matrix-volatility patterns from Leoville and Vigarano show that the matrix has an overall CI-like pattern (Fig. 2A). Peaks of Ba and Eu relative to CI are ascribed to terrestrial weathering (17). In addition, rare earth element (REE) patterns from Leoville and Vigarano matrix are flat and CI-like (*SI Appendix*, Figs. S8–S10), in agreement with previous studies of REEs in Leoville components (17). Moreover, other refractory elements such as Ca and Ti for which chondrule–matrix complementarity is invoked (9, 10) have matrix compositions that do not significantly deviate from CI composition ($Ca = [1.02 \pm 0.5] \times CI$, $Ti = [0.86 \pm 0.5] \times CI$ in this study; ref. 18) and the same average Ca/Ti ratios for chondrules and matrix. While we have not measured Al concentrations here, Ca/Al ratios from Allende and Efremovka (reduced CV) matrix range from subchondritic to superchondritic, whereas their chondrules have CI-like Ca/Al ratios (19). The Ca/Al ratios in the matrix, especially for the less-altered Efremovka, can mostly be attributed to variations in Ca (19), which is highly mobile during secondary alteration. In addition, the distribution of Ca, Al, and Ti in CV chondrite components is dependent on the incorporation of refractory inclusions, and, hence, chondrule–matrix complementarity cannot be considered without contribution from refractory precursors (20). A detailed examination of the CV volatility patterns shows that average matrix and chondrule compositions do not mirror each other but exhibit similar peaks and valleys (*SI Appendix*, Fig. S12A). We interpret this tracking of chondrules and matrix to be caused by terrestrial weathering of the bulk chondrite, since it mainly affects highly mobile elements such as U, Sr, Ba, Rb, and Cs (ref. 21 and *SI Appendix*). This effect is even stronger in patterns displayed by Maribo (Fig. 2C and *SI Appendix*, Fig. S15A), which are nearly identical for chondrules and matrix. Again, the matrix of Maribo shows no evidence for volatile gain and exhibits an overall CI-like pattern, in agreement with presolar grain abundances in CM chondrites (22) and model-dependent deductions of the CM matrix composition (*SI Appendix*). The fact that chondrules from Maribo show very limited volatile loss can be attributed to their relatively small size. During chondrule formation, chondrule melts are suggested to interact with a surrounding volatile-rich gas that will be partially reabsorbed by the chondrules, as evidenced by porphyritic pyroxene chondrules from Vigarano (23) and pyroxene-rich rims from CV chondrules (refs. 24 and 25 and *SI Appendix*). Since CV chondrules are typically an order of magnitude larger than CM chondrules (26), their larger surface area likely caused CM chondrules to reabsorb volatiles to a greater extent. Peaks of U, Sr, and Ba in the chondrule and matrix volatility patterns of Maribo relative to CI chondrites are also observed in bulk compositions of Cold Bokkeveld, Nogoya, and Murchison (27), and we suggest these peaks are caused by terrestrial weathering (*SI Appendix*). In contrast to CV and CM chondrites, the CR chondrite NWA801 shows a mirroring pattern for nonrefractory elements with condensation temperatures $<1,600$ K (Fig. 3 and *SI Appendix*, Fig. S16). The average chondrule composition is more volatile-depleted relative to CV and CM chondrites, and the average matrix is superchondritic and increasingly enriched in volatiles at condensation temperatures $>1,300$ K. Together with the Fe/Mg and Si/Mg data (Fig. 1), the CR chondrites are the only chondrite group that potentially supports some form of complementarity. This would have to be carefully examined

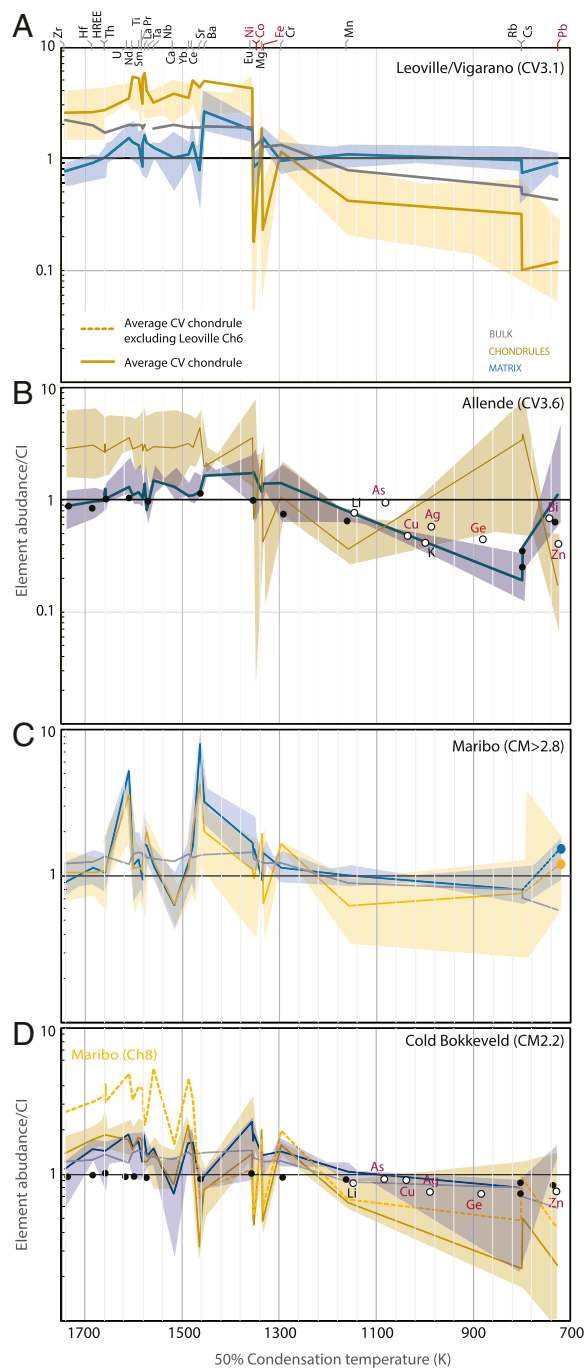


Fig. 2. Element abundance relative to CI composition according to decreasing 50% condensation temperature after (28) for average Leoville and Vigarano (A), Allende (B), Maribo (C), and Cold Bokkeveld (D) chondrules and dust rims and bulk CV and CM composition (16, 29). The shaded areas cover all individual chondrule and matrix locations (*SI Appendix*, Figs. S14 and S15). The CV chondrule average is presented with (solid line) and without (dashed line) an Al-rich chondrule from Leoville. Note that the CV and CM bulk are only an approximate average of chondrules and matrix, since CAIs, metal, and tochilinite–cronstedtite intergrowths are not taken into consideration. B and D include matrix data from ref. 13 (solid black circles are also measured in this study, open circles are not), where in Allende, volatile chalcophile elements (purple) are typically enriched over lithophile elements due to their incorporation in sulfides during secondary alteration. See *SI Appendix* for detailed zoom-ins of refractory plateaus with mirroring chondrule and matrix patterns (*SI Appendix*, Fig. S12). D shows chondrule Ch8 from Maribo, which likely represents a more altered CM clast within Maribo (*SI Appendix*). See yellow dashed line.

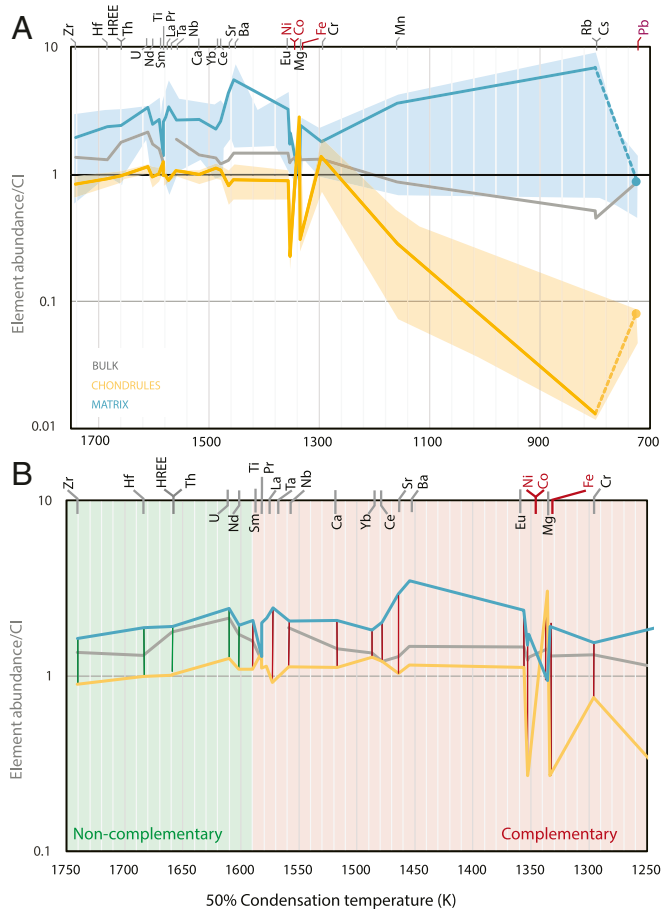


Fig. 3. Element abundance relative to CI composition according to decreasing 50% condensation temperature (28) for average NWA801 chondrules and dust rims and bulk CR composition from ref. 29 (A). (B) Zoom-in of A, showing mirroring (i.e., complementary) and nonmirroring patterns (i.e., noncomplementary) for matrix and chondrules (SI Appendix, Fig. S16).

with a mass balance equation of all elements. For CV and CM chondrites, the Fe/Mg ratios contradict the volatility patterns.

The Effect of Secondary Alteration

We have investigated the volatility patterns of altered and relatively unaltered chondrules and dust rims from CV and CM chondrites to identify the mobilization of elements during thermal metamorphism and aqueous alteration. We have combined the patterns of Leoville and Vigarano to represent average “unaltered” CV chondrite compositions (Fig. 2A), since Vigarano and Leoville chondrules have nearly identical volatility patterns (SI Appendix, Fig. S14A) and the chemical alteration of Vigarano mostly shows within a few tens of microns along the chondrule boundaries (SI Appendix, Fig. S2). Allende, the more altered CV chondrite, shows markedly different patterns for matrix and chondrules at condensation temperatures below the refractory plateau (Fig. 2B), in agreement with previous LA-ICPMS studies of Allende matrix (13). Mobilization is evident for siderophile elements such as Fe, Ni, and Co, which are depleted from the chondrules. Moreover, more volatile elements such as Rb and Cs are enriched in the chondrules but depleted in the matrix relative to the unaltered CV chondrites. Overall, the degree of complementarity between matrix and chondrules increases during thermal metamorphism but decreases for the Fe/Mg ratio (SI Appendix, Fig. S12B). Within the refrac-

tory plateau, some elements mobile in fluids (e.g., Sr and Ba) reflect remobilization, which may be related to crystallization of feldspar in the chondrule mesostasis. In addition, light REEs such as La and Ce, but also Hf and Zr are slightly enriched in the matrix relative to the heavy REE (SI Appendix, Fig. S8). The changes in elemental patterns observed between Maribo and Cold Bokkeveld chondrules and matrix is markedly different compared to those observed in CV chondrites (Fig. 4A and B). This likely reflects the difference between mobilization during aqueous alteration and thermal metamorphism. For example, chondrules, and to lesser extent matrix from Cold Bokkeveld, are enriched in Zr, Hf, REE, and Ca relative to Maribo. These elements are typically enriched in CAIs, suggesting that during aqueous alteration, significant exchange takes place between CAIs and the rest of the chondrite. This mechanism has been previously proposed by ref. 14 to explain amongst others, the W complementarity in Allende (12). However, mobilization of W may have been less extensive during thermal metamorphism (even though assisted by metasomatic fluids; ref. 30), as evidenced by the behavior of similarly refractory elements such as Hf and Zr (Fig. 2 and SI Appendix, Fig. S12). Furthermore, we observe loss of siderophile elements from the chondrules, which to some extent is taken up by the matrix. No significant changes are seen for Mg between chondrules of Maribo and Cold Bokkeveld, showing that the lower Fe/Mg ratios in the latter are the results of Fe loss, rather than Mg enrichment. In contrast, the lower Fe/Mg ratios for Cold Bokkeveld matrix are attributed to an enrichment of Mg, possibly derived from CAIs or perhaps from tochilinite-cronstedtite inclusions. Lastly, Cold Bokkeveld shows significant loss of Sr, Ba, U, Rb, and Pb from the matrix, as well as Sr, Ba, and U from the chondrules. These elements are all highly mobile during aqueous alteration. The fact that changes in these features are clearly less pronounced between altered and unaltered CV chondrites suggests that fluid alterations

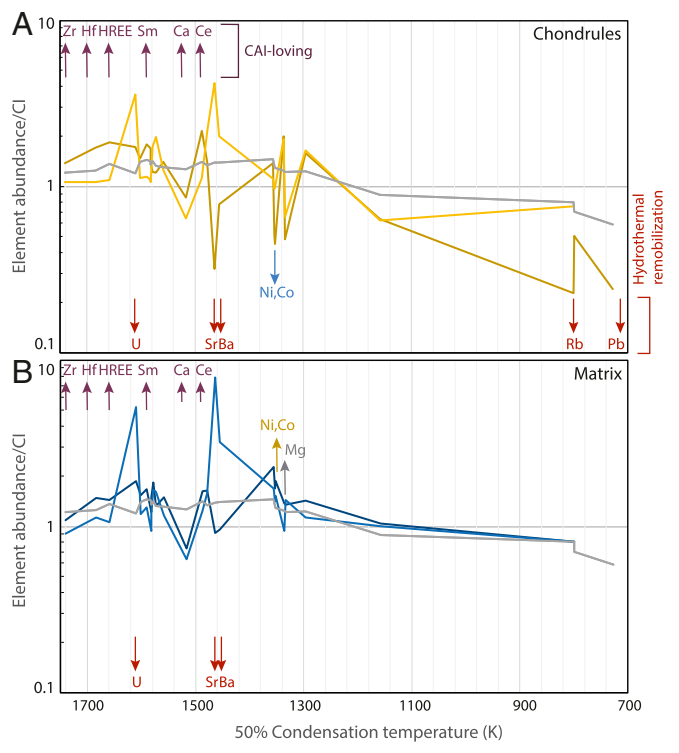


Fig. 4. Element abundances relative to CI composition according to decreasing 50% condensation temperature (28) of CM chondrules and matrix of unaltered and altered samples. (A) Chondrules of Maribo and Cold Bokkeveld compared. (B) Matrix compared.

during thermal metamorphism on the CV parent body were less important.

The absence of an unaltered CR chondrite in this study prevents a thorough assessment of elemental mobilization during aqueous alteration on the CR parent body. We note, however, that NWA801 likely underwent only mild aqueous alteration (*SI Appendix*), implying that the observed volatility patterns are primary features.

A Noncomplementary Unifying Model

We have established that when regarding the most unaltered CM, CV, and CR chondrites as 2-component systems, Fe/Mg ratios of chondrules and matrix appear to be complementary, a trend that is reduced during progressive secondary alteration. The overall matrix composition of CV and CM chondrites appears to be CI-like and does not record reabsorption of volatiles lost during chondrule formation or acts like an elemental mirror to chondrules. This is at odds with the seemingly complementary Fe/Mg and Hf/W ratios and W and Mo isotope

signatures within CM and CV chondrites (8, 11, 12). We propose below a chondrule and matrix accretion model that can explain both datasets and rules out complementarity. Type I (FeO-poor) chondrules are ubiquitous in CV, CR, CM, and ordinary chondrites (>95% of CV, CM, and CR chondrules; ref. 31) and are often “armored,” containing FeNi metal or sulfide coatings on their host chondrule (32, 33). During transient heating events in the disk, chondrule precursors are liquefied and immiscible metal/sulfide (MS) and silicate melts are generated. Centrifugal forces on a rotating chondrule allows for metal–silicate separation through migration of MS melts to the surface (33). Subsequently, this MS either wets the chondrule or recondenses on the chondrule after evaporation (32). We observe these metal/sulfide rims around primary and secondary chondrule cores as well as around igneous rims in our Leville, Vigarano, and NWA801 sections and to a lesser extent in Maribo (*SI Appendix, Fig. S18*). If such armored chondrules were to accrete CI-like dust, then oxidation of the metal during the early stages of aqueous alteration would enrich the surrounding matrix

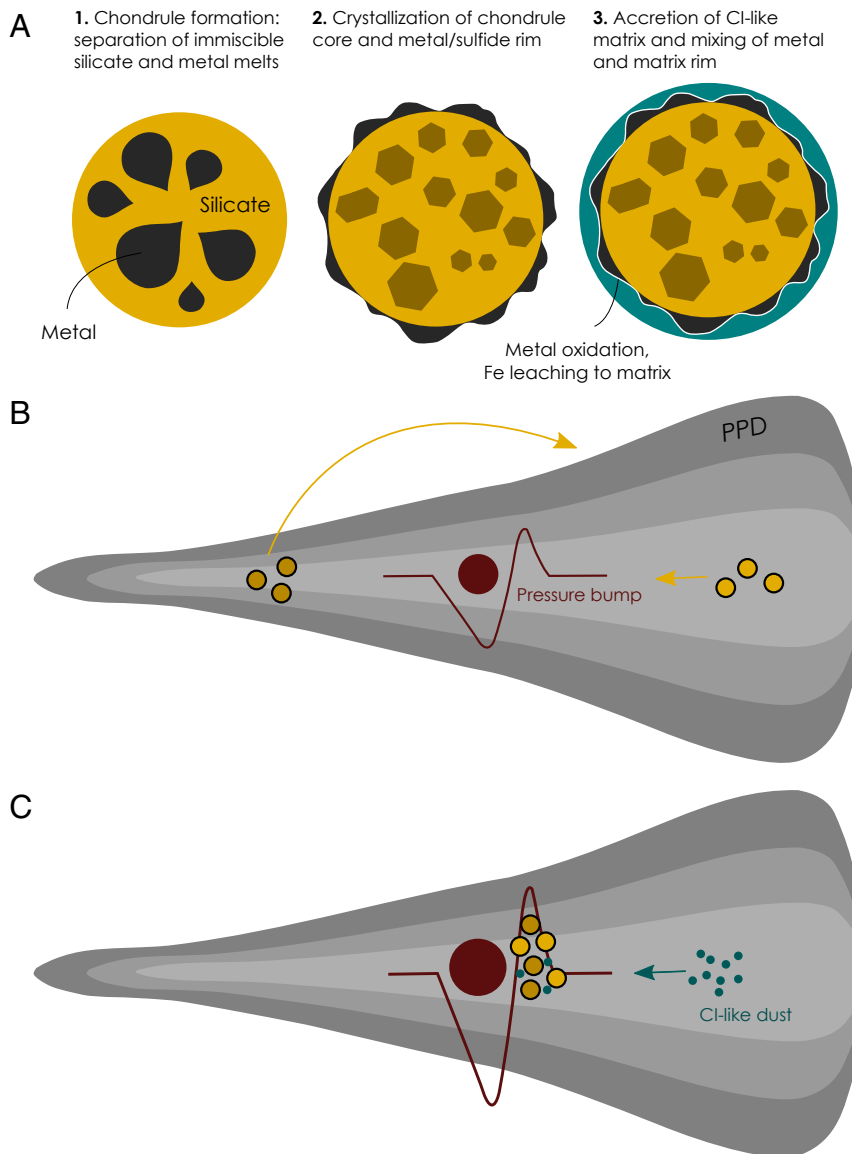


Fig. 5. A unifying model explaining the complementarity paradox. (A) Chondrule formation and accretion of CI-like dust. (B) Chondrule formation and transport in the protoplanetary disk (PPD). (C) Chondrule storage in pressure traps.

in Fe and other siderophile elements (*SI Appendix, Fig. S18H*). Alternatively, physical erosion of the metal rim (*SI Appendix, Fig. S18E*) would yield similar results: the resulting matrix composition would be CI-like but with an increased Fe/Mg ratio complementary to the host chondrule. Moreover, the leaching of Fe to the matrix is accompanied by siderophile elements such as Co, Ni, Mo, and W that are carried by the metal phase (11, 12). Hence, the observed chondrule–matrix complementarity for Fe/Mg ratios and Fe, Mo, and W isotopes can be ascribed to the mixing of metal/sulfide with a CI-like matrix, which is in agreement with mass balance calculations between chondrules, metal, and matrix (*SI Appendix*). As such, the main implication of this study is the absence of genetic relationships between chondrules and matrix in CV and CM chondrites and the existence of an apparent complementarity between these fractions, caused by a third key component, the metal. Another important implication of our observations is that the inferred Hf–W ages of combined chondrule and matrix isochrons from CV chondrites are not valid, since they assume a genetic relationship between these chondritic components (11). Essentially, our model does not require chondrules and matrix from a single chondrite to have formed in the same reservoir but necessitates mass transport in the protoplanetary disk (*Fig. 5B*). This involves the transport of matrix to the chondrule forming region but likely also extends to the transport and storage of individual chondrules throughout the disk, as evidenced by their individual Cr isotope signatures and Pb–Pb ages (3–6). Chondrules could have been stored in loose rubble pile asteroids within pressure traps at planetary gaps (*Fig. 5C* and *ref. 34*), which allow for size sorting of chondrules in various chondrite groups according to the filter efficiency of the dust traps. The increasing efficiency of the dust trap during the growth of a Jupiter-sized planet, would have allowed transport of possibly thermally processed dust, complementary to the chondrules at early times, and accretion of inward migrating CI-like dust at later times within the dust trap. Finally, gravitational collapse of these rubble piles would result in cementing of chondrites containing chondrules alongside each other with varying nucleosynthetic isotope signatures and ages. We note that this model addresses the generic constraints of chondrule and matrix transport within the protoplanetary disk and that a more detailed perspective on the accretion of different chondrite groups with their various chondrule populations requires full knowledge of

their age distribution, isotope systematics, and physical properties (e.g., texture, size, compounds; *ref. 35*). Collectively, this model invokes a generic chondrule-formation process to explain the apparent complementarity between chondrules and matrix in CV and CM carbonaceous chondrites, which may also pertain to ordinary chondrites in which armored chondrules are ubiquitous. Lastly, evidence for complementarity is found between chondrules and matrix of CR chondrites. CR chondrite matrix may be different from that of CM and CV chondrites and could consist of a second dust generation, in agreement with late accretion of CR chondrites. This complementarity is also consistent with the similar Cr and Mg isotope compositions of CR chondrules and bulk (5, 6).

Materials and Methods

We have selected chondrules and surrounding dust rims from CV, CM, and CR chondrites (for a full sample description, see *SI Appendix*). Detailed back-scattered electron images and high-resolution elemental maps were made of these objects by Zeiss EVO MA10 SEM/energy-dispersive X-ray spectroscopy (SEM/EDS) at the Institut de Physique du Globe de Paris (IPGP) (Paris), from which major element concentrations were quantified of regions of interest for laser ablation inductively coupled plasma mass spectrometry (LA-ICPMS) at the Institut Universitaire Européen de la Mer (Brest). We have analyzed 33 major, minor, and trace elements, spanning a range of 50% condensation temperatures between 700 and 1,750 K. LA-ICPMS analyses were performed with a 193-nm Compex Pro102 Coherent Laser Ablation System coupled to an Element XR high-resolution mass spectrometer. Detailed acquisition parameters and data reduction methods are described in *SI Appendix*.

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