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# High-temperature dust formation in carbon-rich astrophysical environments

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+ Extended Data

#### 1 Abstract

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3 Condensation processes, responsible for the main chemical differences between gas and solids 4 in the galaxy, are the major mechanisms that control the cycle of dust from evolved stars to planetary systems. However, they are still poorly understood, mainly because thermodynamics 5 6 and kinetic models of nucleation or grain growth are lacking experimental data. To bridge this gap, we used a large volume three-phase AC plasma torch to obtain a full high temperature 7 condensation sequence at elevated C/O ratio from a fluxed chondritic gas composition. We 8 show that the crystallized suites of carbides, silicides, nitrides, sulfides, oxides and silicates and 9 the bulk composition of the condensates are properly modelled by a kinetically inhibited 10 condensation scenario controlled by the gas flow. This validates for the first-time 11 thermodynamic predictions of condensation sequence at high C/O ratio. On this basis and using 12 13 appropriate optical properties, we also demonstrate the influence of pressure on the dust chemistry as well as the low probability of forming and detecting iron silicides in asymptotic 14 giant branch (AGB) circumstellar environments as well as in our chondritic meteorites. 15

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# 19 Introduction

21 Cosmic dust is of crucial importance for the evolution of our galaxy since it actively participates 22 in the cycle of matter (gas and dust) from the interstellar medium (ISM) to stars and back from 23 stars to the ISM<sup>1</sup>. Dust grains form principally in the gas outflowing from evolved stars and in 24 the ejecta resulting from supernova (SN) explosions. Amongst progenitors, AGB stars are 25 assumed to be the major contributor to the global cosmic dust budget<sup>2</sup>. During their late evolutionary stages, low- and intermediate-mass stars (< 8 M<sub>☉</sub>) become red giants, increasing 26 their radius by 2-3 orders of magnitude, and decreasing their surface temperature to 2000-3000 27 K. Further evolution of these stars (AGB phase) lead to significant mass loss of gaseous 28 29 molecules and dust grains giving rise to circumstellar envelopes (CSE). Mass-loss rates of AGB 30 stars, determined with various observational methods, are typically in the range of  $10^{-8}$ – $10^{-5}$  $M_{\odot}$ . year<sup>-1</sup> but values as high as  $10^{-4} M_{\odot}$ . year<sup>-1</sup> have been found for more extreme objects<sup>3</sup>. 31 32 Around cool stars, dust is formed in dense shells created by stellar pulsations or large-scale convective motions which propagate gas beyond the stellar surface<sup>4</sup> where gas cooling yields 33 dust condensation. These dust grains are then accelerated away from the star by radiation 34 pressure, dragging the gas away through friction, creating outflows with typical velocities of 35 5–30 km/s and eventually feeding the ISM<sup>5</sup> contributing thus to the material out of which our 36 37 solar system formed.

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39 Dust forms from the matter already present when the stars formed and from the elements 40 produced by the stars themselves. The dust composition changes as the star evolves and notably when the carbon to oxygen ratio changes during the episodic third dredge-up process<sup>6</sup>. 41 Thermodynamic equilibrium calculations have provided convincing explanations of the marked 42 chemical differentiation between oxygen-rich and carbon-rich AGB stars based on the high 43 44 bond energy of carbon (i.e., 1077 kJ.mo $l^{-1} \approx 11.1 \text{ eV}^7$ ) and that CO is a very stable gaseous 45 molecule at relatively high temperatures. Consequently, the dust chemistry in the inner CSE of AGB stars,  $R = 1 - 10R_*$  where  $R_*$  is the stellar radius, is primarily controlled by the carbon-to-46 oxygen ratio. When C/O  $\leq 1$  (O-rich stars), all carbon is bound in the CO molecule and the 47 48 oxygen in excess participates to oxygen-rich dust formation<sup>8</sup>. In contrast, when  $C/O \ge 1$  (C-rich stars) carbon is in excess while all the oxygen is bound in CO and hydrocarbons molecules and 49 carbon-rich grains are formed<sup>9,10</sup>. While the C/O ratio likely explains the fundamental 50

difference between C-rich and O-rich stars, temperature and pressure within their inner
 circumstellar envelopes (CSEs) play obviously a significant role (see below).

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Complex dynamical effect in stellar outflows<sup>3</sup> and highly diverse circumstellar O-rich and C-54 rich grains inventory in chondrite meteorites<sup>11,12</sup> highlight, however, the need to consider 55 56 beyond simple thermodynamic equilibrium in order to understand dust formation around AGB stars, including kinetics, nucleation/seeding processes and models like chaotic solid 57 formation<sup>8,13, 14,15</sup>. This issue has been already the subject of extensive theoretical works<sup>13</sup>. Ref<sup>16</sup> 58 59 recently re-emphasized the importance of kinetics for dust formation in astrophysical environments, and the need for laboratory condensation experiments to provide insights into 60 the formation of dust and the reactions that allow their growth. While nucleation and growth of 61 simple metal or H<sub>2</sub>O grains from a gas have been extensively studied for applications in 62 atmospheric or material science<sup>17,18,19,20,21</sup>, condensation of more complex gas, such as those of 63 stellar outflows, is less well studied. The difficulty resides in simulating astrophysical 64 conditions in the lab: (1) producing a multi-elemental refractory gas of controlled composition 65 66 and (2) condensing it in equilibrium/disequilibrium conditions at very high temperature<sup>22,23,24,25</sup>. Several conditions need indeed to be met for dust/condensed matter formation, such as densities 67 above a certain threshold to ensure sufficient collisions between the constituent particles, 68 69 moderate temperatures below the stability threshold of the dust component, and sufficient time 70 for the nucleation and growth of molecular clusters into larger grains.

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72 Here 600 g of a 20 µm-sized powder of NWA869 ordinary chondrite have been processed for an hour inside a large volume 250 kW three-phase alternating current (AC) plasma torch with 73 74 hot graphite electrodes operating at PERSEE, Mines-Paris-Tech, Sophia-Antipolis, France (Fig. 75 1). Chondritic dust particles were injected using argon gas for propulsion into a 5000 K H<sub>2</sub>-Ar admixture gas plasma formed between three graphite electrodes (see Methods). The vapor 76 formed in the plasma flew downwards in a cylindrical meter-sized graphite-coated chamber in 77 78 which condensation occurred according to the thermal gradient at a pressure  $\approx 10^5$  Pa. Nitrogen 79 was injected during the run to avoid hydrogen flammability. The experiment was terminated by 80 shutting down both the electrical power and the powder supply.

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**Fig. 1:** a) 240kW three electrode arc plasma torch (PERSEE, Mines ParisTech, Sofia-Antipolis, Nice) in which 600 g of a 20 µm-sized powder of NWA869 ordinary chondrite have been processed for an hour. b) Simulated 3D thermal regime inside the condensation chamber using the ANSYS Fluent computational fluid dynamic software. A removable Tshape graphite rod was placed at the center of the chamber and was recovered after the experiment for chemical and mineralogical analyses of the condensates. The 3D simulation was performed in a computational domain that does not explicitly incorporate the T-shape graphite rod. To account for the rod's presence, the torch power was set to the experimentally determined value, maintaining the appropriate source temperature while ensuring overall energy balance. To achieve this, losses induced by the rod were redistributed onto the walls of the chamber, preserving the global thermal behavior of the gas flow. While the thermal distribution may not be locally exact near the rod location, the rod temperature can be considered to lie within the thermal boundary layer that forms near the walls. Equivalence calculations suggest that the rod temperature is approximately between 1/8 and 1/6 of the nozzle diameter away from the walls.

100 A removable T-shape graphite rod was placed at the center of the chamber and was recovered 101 after the experiment for chemical and mineralogical analyses of the condensates (Fig. 1 and 102 Extended Data Fig. 1). Even if this experiment remains far from CSE conditions, e.g.,  $P << 10^2$ 103 Pa, which are technically impossible to achieve at present, we show in the following how such 104 high temperature condensates resulting from fluxed chondritic gas at atmospheric pressure in a 105 severe thermal gradient help us to put stringent constraints on dust formation in stellar outflows. 106



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110 111 **Fig. 2:** Photographs of the condensates on T-shape graphite rod after experiment. Notice the change of the structure (e.g., wetting, flaking) and the grain size of the condensates from top (high temperature) to bottom (low temperature). See text.

#### 112 113 **Results**

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Owing to this large volume device, the recovery of several hundred grams of high temperature 115 116 condensates obtained at high C/O ratio is a première. The 3D thermal regime inside the chamber as well as the trajectory and velocity of fictive particles (Fig. 1 and Extended Data Fig. 1) have 117 been obtained using the ANSYS Fluent computational fluid dynamics software (see 118 Methods<sup>26</sup>). While condensation occurred in the whole condensation chamber, the T-shape 119 120 graphite rod holder offers enough material (Fig. 2) to detail the chemistry (Extended Data Table 2) and the mineralogy (Extended Data Table 1) of the condensates between 2400 K to 1300 K 121 at ambient pressure and C/O ratio  $\approx 0.93$ . The high C/O ratio is due to and regulated by the 122 123 consumption of graphite from the 3 electrodes during the experiment (see Methods and 124 discussion below).

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126 X-Ray diffraction (XRD) pattern and secondary electron (SE)- and back scattered electron

- 127 (BSE)-Scanning Electron Microscopy (SEM) characterization reveal that condensates obtained 128 at the highest temperatures, i.e., >2050 K, are dominated by iron silicides ( $Fe_xSi_y$  with x/y
- atomic ratio  $\approx 1.2$ ). They formed millimeter-sized droplets wetting the T-shape graphite rod

(Fig. 2). Well crystallized SiC are bathed in the silicides together with rarer and smaller crystals
of Al<sub>4</sub>SiC<sub>4</sub> and TiC (Fig. 3 and Extended Data Fig. 3).

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**Fig. 3:** Representative scanning electron microscope images of high temperature condensates obtained at high C/O ratio. a) Well crystallized SiC bathed in molten FeSi (now solid) from sample #13 condensed at temperature around 2150K. b) Overlapped X-ray elemental map in Ca (pink), Fe (green), Si (blue) and S K $\alpha$  (red) and SE map of # 8 condensates obtained at temperature 1700K showing an association of oldhamite (CaS-MgS), Mg-rich olivine (Mg<sub>2</sub>SiO<sub>4</sub>) and molten droplets of iron silicide (FeSi).

142 A temperature of about 2050 K marks the onset of condensation of oxygen-bearing and sulfur-143 bearing phases with the occurrence of calcium-aluminate (silica poor) glass and of CaS-MgS 144 oldhamite, respectively. Calcium-aluminates, that are initially molten, wet the immiscible FeSi 145 molten droplets and most of the already condensed crystalline phases, including SiC. Oldhamite 146 condenses first as Ca-rich phase and shifts to an MgS-rich composition as temperature drops. 147 AIN (some of which are whiskers) and TiN nitrides appear below on the T-rod in a narrow interval of temperature between 2000-1900 K. In the same range of temperature, olivine 148 149 Mg2SiO4 starts to crystallize from the melt (now glassy and enriched in MgO and SiO2) in close association with FeSi and SiC. Below a temperature range of 1850-1800 K, X-ray diffraction 150 analysis reveals a critical shift in the condensate composition. Graphite becomes the dominant 151 phase, while carbides are no longer detectable. Low Ca-pyroxene MgSiO<sub>3</sub> starts to condense 152 153 below 1700 K forming remarkable 1-D whiskers by a FeSi catalyst-assisted VLS (vapor-liquid-154 solid) growth process as shown in the Extended Data Fig. 3. Iron silicides, Fe<sub>x</sub>Si<sub>y</sub> have x/y 155 atomic ratio close to 2 in these low temperature condensates. Crystals of periclase MgO have also been found in the lowest temperature part of the condensates. Finally, a clear decrease by 156 157 one order of magnitude in the average size of condensed phases from tens of microns to tens of 158 nanometers is noticeable as condensation temperature drops from top to bottom of the T-shape 159 graphite rod.

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Extended Data Table 1 shows the bulk chemistry (Si, Ti, Al, Ca, Mg, Fe, Mn, Na, K, P, C and S) of both the starting NWA 869 chondrite composition as well as the 14 aliquots of condensates sampled on T-shape graphite rod holder measured by Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES).



165

166Fig. 4: Composition patterns for the main major and minor elements of the condensates167considering a kinetically limited condensation with various f values for the phases (see168text). The solid lines correspond to the modeled compositions and the dashed lines to the169measured ones.

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As condensation proceeds downward in the chamber, condensates are first enriched in Fe, Si, Ti and C, then become enriched in Al, Ca, S, and next in Mg, a change in chemistry in good agreement with their mineralogy (Fig. 4). Sulfur is however enriched in the low temperature condensates (< 1600 K). Only trace amounts of Na are found in the lowest temperature condensates while K is totally absent of this high C/O ratio condensation sequence.

# 177 Discussion

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# A. Laboratory condensation of chondritic gas at high C/O ratios

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181 Our results show a sequence of carbides, sulfides, nitrides, and oxides that are generally 182 consistent with calculations made in previous studies that correspond to the low-pressure 183 conditions of protoplanetary disks or stellar atmospheres at high C/O ratios<sup>14,27,28,29,30</sup>. 184 However, there are some differences due to the duration and the high-pressure conditions ( $\approx$ 185 10<sup>5</sup> Pa) of the experiment. Carbon in our experimental condensates is mainly in the form of 186 carbides at high temperatures (above 1850 K) and transitions to graphite at lower temperature.

187 No evidence of amorphous carbon has been found. The most important difference is the stability of liquids in these conditions, namely iron silicide metallic liquid and oxide/silicate liquid, 188 189 which are immiscible with each other. As is well known in materials science, the liquid acting as a catalyst promotes mass transfer in the VLS system and controls the growth and shape of 190 the crystals. The large SiC crystals that crystallize from FeSi liquid<sup>31</sup> and the various 1D 191 192 crystals/whiskers of AIN, MgSiO<sub>3</sub> catalyzed by liquid FeSi droplets on their tips provide 193 convincing examples of the VLS process and its importance at high-pressure condensation 194 conditions (Fig. 3 and Extended Data Fig. 3).

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196 Regarding the chemistry of condensates (Extended Table 1), the calculated compositions based on thermodynamic equilibrium or fractional condensation do not match the measured 197 198 compositions, while the calculated mineralogical sequences do (see Methods). In other words, this suggests that an equilibrium condensation sequence, i.e., the order of phase appearance 199 200 during condensation, is not a good discriminant for identifying the exact process of 201 condensation (see below). We thus explore a partial fractional condensation model (Extended 202 Data Fig. 4) where only a fraction f of what was expected to condense in equilibrium could 203 effectively condense due to kinetic limitations (See Methods).

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205 The condensation pattern of Si, Fe, Al and Ca determined with the model fits well with the 206 experiments (Fig. 4). Fe concentrations in the condensates remain high (~ 60 %) until 1950 K, 207 when phases other than FeSi start condensing (SiC, AlN, CaS, oxide melt). The modelled x/y 208 ratio of the Fe<sub>x</sub>Si<sub>y</sub> molten alloy ranges from 1 to about 2 as the temperature decreases, which is perfectly consistent with the measured compositions. The Si concentration remains almost 209 210 constant along the rod, showing a roughly constant condensation of this element in various 211 phases: FeSi, SiC, oxide melt, followed by silicates. Ca concentration shows a sharp increase until CaS becomes stable (Fig. 4). Its concentration increases until it reaches a maximum at 212 1900 K. At this temperature, CaS is no longer stable, and Ca is mainly partitioned in the oxide 213 214 melt. Al concentrations show a rather similar pattern, with a sharp increase when AIN becomes 215 stable, followed by a short decrease until it reaches a plateau at 1900 K when AlN is no longer stable, and Al condenses in the oxide melt. Both Ca and Al concentrations sharply decrease 216 217 after 1800 K when Ca-poor and Al-poor phases start condensing (i.e., olivine and periclase). 218

219 Despite the overall good agreement, some discrepancies between the model and the experiment can be observed (Fig. 4). The main issue is related to sulfur which has very low predicted 220 221 concentration in the low-temperature sections compared with the measured concentrations. The 222 main S-bearing phase in the thermodynamic simulations is CaS, while the experimental results are not consistent with the calculated condensation curve for this species: to obtain a CaS phase, 223 224 the concentration of Ca should have been much higher than what is observed. Furthermore, no 225 new sulfide phase was identified in the low-temperature segments of the rod with XRD. There 226 is thus a clear problem with the sulfur data that could have arisen for several reasons including 227 i) analytical artifact with the measurement, ii) contamination by the condensation chamber, and iii) thermodynamic underestimation of the amount of S that could get dissolved in the FeSi, 228 and/or iv) significant solubility of sulfur in silicate liquids under reducing conditions<sup>32</sup>. Despite 229 230 this difference in concentrations, the predicted and observed patterns of S concentrations are very similar, with a first peak of concentration at 1950 K corresponding to the initial stability 231 232 field of CaS and a sharp increase until a plateau around 1750 K corresponding to the second 233 stability field of CaS.

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235 Our model, which incorporates a kinetically limited condensation mechanism, effectively 236 reproduces the experimental results for most major elements at high C/O ratios. This provides the first experimental validation of thermodynamic calculations employed to predict condensation sequences under such conditions. Notably, it is also established that the kinetically inhibited condensation sequence does not differ from the equilibrium sequence, whereas the relative abundances of the condensed phases and the resulting bulk composition do. Consequently, the bulk composition, rather than the mineralogical sequence of condensation, serves as a more reliable indicator of the extent of kinetic limitations during the condensation process.

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Pressure versus C/O ratio diagram showing the stability field of first condensates Fig. 5: using the starting bulk composition of the solar composition from ref<sup>33</sup>, except for the C 247 abundance. In this range of P and C/O ratio, Ti<sub>2</sub>O<sub>3</sub> and CaTiO<sub>3</sub> are stable at low C/O ratio; 248 note that Al<sub>2</sub>O<sub>3</sub> (not shown) is stable at lower C/O ratios<sup>28</sup>. At higher C/O ratio, oxides are 249 250 no longer stable, and TiC and graphite are becoming predominant. Red square corresponds 251 to our experimental conditions. Thin lines stand for the  $C_2H_2/CO$  ratio in the gas phase at 252 the saturation of the first condensates, i.e., Ti<sub>2</sub>O<sub>3</sub>, CaTiO<sub>3</sub>, TiC or graphite - depending on 253 the C/O ratio and the pressure. Notice the significant pressure influence of the transition 254 between carbon- and oxygen-bearing phases (red curve). As pressure approaches  $10^5$  Pa, 255 the C/O threshold drops well below  $\approx 1$ , indicating a strong destabilization of oxide phases 256 in favor of carbides (See text). 257

It is crucial to highlight that while the traditional distinction between O-rich and C-rich dust 258 259 chemistry commonly aligns with a C/O value of unity, the C/O ratio derived from our C-rich dust-generating experiment dips below this threshold, settling at C/O = 0.93. This reported ratio 260 is thus typical of a S-type star rather than that of a C-type star. According to previous 261 thermodynamic calculations<sup>28</sup>, the transition between reduced phases (e.g., carbides, nitrides) 262 and oxygen-bearing phases (e.g., oxides, silicates) is also observed at a C/O ratio of 263 approximately 0.96 at a total pressure of  $10^2$  Pa, indicating a significant pressure influence on 264 265 this transition. This is corroborated by our thermodynamic calculations, as shown in Figure 5. Our results confirm a significant pressure-dependent shift in the C/O value required for oxide 266 267 formation. As pressure approaches 10<sup>5</sup> Pa, C/O threshold drops well below  $\approx$  1, indicating a strong destabilization of oxide phases in favor of the carbides. This is concomitant to the 268

dramatic increase in the proportion of carbon-hydrogen bond as pressure rises, as indicated by the increase of  $C_2H_2/CO$  ratio of the vapor phase (Fig. 5). The observed preference for C-H bond formation can be attributed to the inherent weakness of this bond compared to the C-O bond in carbon monoxide. The C-H bond energy is several hundreds of kJ/mol lower, making it significantly easier to break and participate in the formation of new reactive carbon species in the vapor phase under pressure. Such pressure effect can be rationalized by a reaction involving the main gas species of the type:

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$$2 CO_{(g)} + 3 H_{2(g)} = C_2 H_{2(g)} + 2 H_2 O_{(g)}$$

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which, according to Le Chatelier's principle, will be shifted towards the product side ( $C_2H_2$ formation) as pressure increases (P $\Delta$ V negative). This aligns perfectly with the observed increasing stability of C-bearing condensed phases at higher pressures and with the presented experimental data (Fig. 5). In light of this, our condensation sequence at 10<sup>5</sup> Pa with C/O = 0.93 replicates the sequence observed in CSE with higher C/O ratios. In what follows, we explore the consequence of our thermodynamic calculations extended to low-pressure conditions that are representative of stellar atmospheres with high C/O ratio such as AGB S- and C-stars.

# 287 **B.** Condensation in C-rich stellar atmospheres

Evolution of AGB stars is controlled by two main processes: dredge-ups that bring material 288 from the inner shell to the most outer layers, thus increasing the C/O ratio of the stellar 289 atmosphere<sup>34,35</sup>, and mass loss including C through stellar winds<sup>36</sup>. Dust formation by 290 condensation is crucial since stellar winds are driven by momentum transfer from the photons 291 emitted by the star to the surrounding  $dust^{3,37}$ . The accelerated dust then drags the surrounding 292 293 gas and stellar winds are formed. The acceleration of dust by radiative pressure depends on the 294 efficiency of the momentum transfer from the photons to the grains, namely the mean opacity 295 of the dust. Stellar winds can only form if the radiative pressure force exceeds the local gravity 296 force of the star, which is when the mean opacity of the dust exceeds a critical value that 297 depends on the star mass and luminosity. However, momentum transfer is not the only light-298 matter interaction that plays an essential role. In particular, the light absorption properties of some minerals can heat up the grains such that the actual temperature of the dust grains can be 299 300 different from the local gas temperature. Thus, these grains can be heavily heated up and their 301 temperature can then exceed their condensation temperature. Consequently, their condensation can be inhibited, destabilizing these minerals even though the gas temperature should allow 302 condensation. This phenomenon has already been described for Fe-bearing olivines around 303 AGB M-stars<sup>38</sup>. This effect will contribute to increasing the effective oversaturation of this 304 305 phase. As the gas and dust flow away from the star, the temperature and pressure decrease such that at some point the predicted dust temperature shall correspond to its condensation 306 temperature<sup>39,40</sup>. The evolution of the AGB stars critically depends on the properties of the 307 condensing grains with respect to light-matter interactions. According to ref<sup>39</sup>, the degree of 308 heating of a mineral phase will depend on a parameter p whose value can be calculated if the 309 dependence of the coefficient of absorption  $Q_{abs}$  as a function of wavelength is known. The 310 values of p for mineral phases predicted in our condensation sequence were calculated based 311 on their optical properties (whenever they were not directly available from the literature) and 312 313 are reported in Extended Data Table 4. The p values were used to compute the actual temperature of the dust as a function of the distance from the star<sup>40</sup> (See Methods). 314



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Fig. 6: a) Comparison of the thermodynamic condensation temperature (solid line) and the 317 dust temperature (dashed line) as function of the distance from an AGB star with a C/O 318 ratio of 0.93 for six different minerals. The composition of the gas and the pressure profile are from ref<sup>14</sup>. The surface temperature of the star is considered to be 2500 K. Condensation 319 320 of minerals can only occur when the actual dust temperature is lower than the condensation temperature, i.e the minimum condensation distance is the intersection of the dashed line 321 322 and the solid line. b) Condensation distances for various mineral phases calculated for C/O 323 of 0.93, 1 and 1.5. The distances are expressed as multiples of the star radius and were 324 obtained using the same approach as in a).

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326 The condensation temperatures of dust with a C-star composition were then calculated using 327 the same thermodynamic database as that used for modeling our experimental condensation sequence. The starting bulk composition was the solar abundance from ref<sup>33</sup> with an increased 328 329 C abundance. The distance where a mineral first condenses was calculated for a temperature

and pressure profile in the extended atmosphere of a representative AGB star<sup>14</sup>. In this 330 calculation, the abundance of C was increased over the solar value starting from a value of 0.93 331 332 (corresponding to our experiment) up to 1.5, since most C-rich AGB stars have 1 < C/O < 1.5. The results of these calculations were then plotted in Figure 6 where dust temperatures are also 333 reported. In principle, the minimum distance where dust can condensate is located at the 334 335 intersection of the condensation temperature and dust temperature curves. For TiC, SiC and 336 olivine, the distance is close to the star ( $\langle 2R_*\rangle$ ). In these conditions, the pressure is high enough (>10<sup>-2</sup> Pa) for condensation to occur. However, for CaS, FeSi and C, the condensation can only 337 take place at distances greater than  $5R_*$ . At this distance, the pressure is lower than  $10^{-5}$  Pa and 338 the condensation must take place at a much lower rate owing to a lower local density. In order 339 to further determine whether the formation of FeSi dust could take place despite the low density 340 at such a large distance from the star, we evaluated the timescale necessary for FeSi dust growth 341 342 compared with the timescale of the outflow itself using the approach described in ref<sup>41</sup> using an appropriate set of parameters (See Methods). The results are shown in Extended Data Fig. 5. 343 The growth timescale for dust particles between  $10^{-1}$  and  $10^{-3}$  µm is always an order of 344 magnitude longer than the outflow timescale. This shows that provided the dust temperatures 345 prevent condensation at a close distance from the star (here >5R\*), the low density in this 346 347 environment then becomes prohibitive for allowing a rapid enough grain growth. Consequently, 348 our kinetic models predict that FeSi is unlikely to form in the outflows of C-rich AGB stars, 349 despite thermodynamic predictions favoring its condensation under these conditions. In addition, one could note that the detection of IR signature for FeSi in stellar outflows is itself 350 351 difficult<sup>42</sup> due to the lack of distinct spectral features.

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Assuming that our calculations capture the essence of processing affecting dust generated in C-353 354 rich AGB stars, these results can be compared with direct observations of AGB stellar outflows and presolar grains preserved in primitive chondrites. Mid-infrared (MIR) survey of evolved 355 stars has revealed a clear dichotomy in the chemical properties of dusty outflows. Carbon dust, 356 357 be it amorphous carbon or crystallized graphite<sup>43</sup>, and silicon carbide appear together in the 358 outflows of C-enriched stars, whereas silicates, alumina and water ice dominate the dust emission of stars with C/O<<144. C-rich dust, such as graphite, SiC and polycyclic aromatic 359 360 hydrocarbons (PAHs) do show emission features at 3-30 µm. The carriers of the 21 µm and 30 361 um features in the outflows of C-rich stars are currently still debated<sup>45</sup>. For instance, the feature observed around 30 µm in the outflows of C-stars is often attributed to MgS but its formation 362 is unclear<sup>45,46,47</sup>. An important feature is that AGB dust has an extremely low Fe content, 363 364 although iron is significantly depleted from the gas phase in both circumstellar and interstellar environments and ~90% of iron produced by stars should be in dust grains. These features seem 365 consistent with our modeling, suggesting the absence of FeSi condensation due to kinetic 366 367 constraints. 368

Similarly, the mineralogy and composition of presolar grains provide some strong hints about 369 their birth environment. It is commonly considered that the majority of these grains come from 370 C-rich AGB with abundant SiC<sup>48</sup> and rare C<sup>49</sup>, CaS and MgS and TiC (but also (Zr,Mo)C has also been documented as tiny grains<sup>50,51,52,53</sup>). In contrast, the presence of FeSi is very rare and 371 372 373 only mentioned as a small inclusion by ref<sup>53</sup>. Thus, to a first order, our modeling is consistent with observations in presolar grains. In some cases, TiC has been described as occurring as 374 inclusions inside SiC or graphite dust grains and it has been suggested in previous studies that 375 TiC (or ZrC) could have acted as a seed for heterogeneous nucleation 51,54,55,56,57,58. This could 376 indeed have been the case when they occur in a central position. However, according to the 377 observations of ref<sup>53</sup>, their elongated shape seems to preclude a condensation origin for the TiC 378 inclusions and would suggest also a secondary exsolution. 379

### 380 Conclusion

381

382 Overall, our experiment validates the mineralogical sequence that had been predicted by equilibrium thermodynamic calculations and shows that under kinetic conditions where 383 condensation is limited by flow rate, the equilibrium sequence of minerals is preserved, while 384 385 the phase abundance is under kinetic control. By including the effect of light interaction with minerals that have strong IR absorption, our results also show that one can effectively predict 386 the observed minerals found in C-rich stars, but also in presolar grains. It should be mentioned, 387 however, that wind characteristics of AGB stars could be significantly affected in the case of 388 binary systems with closed-by companions which often have an equatorial density 389 enhancement, accretion disk and complex inner wind dynamics<sup>59</sup>. Nevertheless, this study 390 391 suggests that more extensive condensation experiments such as that presented here could 392 provide novel insights into dust formation in astrophysical environments.

- 393394 Methods
- 395 395 396

# A. Experimental approach

397 The experimental work was performed at Mines ParisTech-PERSEE, Sophia-Antipolis, Nice (Fig. 1). The experimental setup was made of a large volume plasma torch at atmospheric 398 399 pressure. A finely ground powder (~ 20 µm) of the NWA 869 ordinary chondrite (bulk 400 composition given Extended Data Table 1) was injected in a 5000 K Ar-H<sub>2</sub> plasma formed 401 between 3 graphite electrodes. The total volume of plasma thus formed was about 1 L. Electrode erosion enriched the vapor with carbon, driving a constant C/O ratio near unity. This ratio is 402 403 significantly higher than the initial C/O ratio of the starting chondritic material. The vapor formed in the plasma flew downwards in a graphite-coated chamber along which condensation 404 occurred in a temperature gradient. A removable T-shaped graphite rod positioned at the 405 chamber's center facilitated the retrieval of condensates formed due to the chamber's thermal 406 407 gradient. The temperature and flow velocity were determined numerically. The magnetohydrodynamic equations of the gas as well as the thermo-mechanical evolution of 2000 virtual 408 particles were solved using ANSYS Fluent software<sup>26</sup>. From this, we were able to compute the 409 410 temperature field inside the condensation chamber (Fig 1.b.) and the gas flow streamlines. The 411 flow velocity ranged between 1 m/s and 0.6 m/s at the top and end of the chamber respectively, while the temperature ranged from 2400 K to 1300 K at the top and end of the graphite rod, 412 413 respectively. The rod was recovered after the experiment and underwent chemical and mineralogical analyses. Condensates on the T-shaped graphite rod were collected using a 414 scraping technique from fourteen, 5 cm long segments along its length (Extended Data Fig. 2) 415 and Extended Data Fig. 3). The chemical and mineralogical compositions of each segment were 416 then analyzed and are summarized in Extended Data Table 1 and Extended Data Table 2, 417 418 respectively.

419

# 420 **B.** Thermodynamic simulations

421 The thermodynamic simulations of the system were done with the FactSage software. This 422 software computes the equilibrium composition of a system based on Gibbs energy 423 minimization method, given a composition and the P-T conditions of the system. We used the 424 FactPS, FTStel and FToxid databases<sup>60</sup>. By calculating equilibrium phase composition as a 425 function of decreasing temperature, the software is able to simulate condensation starting from 426 a vapor with a chondritic composition, mixed with Ar, N<sub>2</sub>, H<sub>2</sub> and additional C.

427

The system was composed of 600 g of the NWA 869 composition (Extended Data Table 1) and an amount of Ar, H<sub>2</sub> and N<sub>2</sub> that correspond to the total amount of gas flowing in the plasma torch during the experiment. As the actual C/O ratio of the system could not be easily determined, several simulations were performed with a C/O molar ratio ranging from 0.72 to 1.15. For each simulated composition, the pressure was kept at the constant value of 10<sup>5</sup> Pa whereas the temperature ranged between 2700 and 1300 K with temperature steps of 50 K.

434

435 For each temperature step, the software computed the expected phases in equilibrium, including 436 their composition and total masses. We considered three possible condensation behaviors: an equilibrium condensation where the bulk composition of the system remained constant along 437 438 its path, a fractional condensation where all the condensed material was subtracted from the vapor composition at each temperature step and a partially fractional condensation where only 439 440 a fraction f of what was supposed to condense at equilibrium could effectively condense due to kinetic effects (see section C. below). For each of these scenarios, we were able to define a 441 442 condensation sequence and the bulk chemical pattern of the condensates with decreasing 443 temperature. These results were then compared with the actual sequence and the chemical pattern measured on the T-shape rod. 444

# 445446 C. Thermo-kinetic model of condensation

447 Since the data could not be explained by an equilibrium model or a fractional condensation 448 model, we designed a kinetic model that considers the kinetics of condensation as the vapor is flowing through the condensation chamber. The schematic geometry of the experiment is 449 450 presented in Extended Data Fig. 3. The system is composed of the condensation chamber with 451 a radius r = 7 cm. At the center of the chamber, the rod is assumed to be a cylinder with a radius 452 r' = 1 cm. Only a fraction f of what was expected to condense in equilibrium could effectively 453 condense due to kinetic limitations. To evaluate a value of f that would be consistent with 454 kinetic constraints, we included a model for gas flow through the graphite-coated chamber. We 455 considered that the kinetics of condensation was controlled by gas flow, thus neglecting the 456 kinetics of nucleation. The vapor was assumed to flow downward through a vertical cylinder of diameter r. If one considers a slice of thickness dz through the cylindrical tank located below 457 458 the plasma, one can write a mass balance equation for the fluxes (Extended Data Fig. 4): 459

460

$$F_{in}(z) = F_{out}(z+dz) + F_{cond}(z) \quad (1)$$

461 where  $F_{in}$  is the advective flux of gas entering at position z,  $F_{out}$  is the advective flux of gas 462 leaving at position z+dz and  $F_{cond}$  is the condensation flux on the chamber's inner surface. 463

464 One can then write  $F_{in}$  and  $F_{out}$  as a function of the molecular densities  $\rho(z)$  and  $\rho(z+dz)$ 465 respectively:

$$F_{in}(z) = \rho(z)Sv$$

466 467

470 *S* is the surface of an axial section through the system, i.e.,  $S = 2\pi (r^2 - r'^2)$ , v is the mean vertical 471 gas velocity. The condensation of elements follows the Hertz-Knudsen equation:

 $F_{out}(z+dz) = \rho(z+dz)Sv$ 

472 
$$F_{cond}(z) = \frac{S'\alpha(P_i(z) - P_{i,sat}(z))}{\sqrt{2\pi MRT(z)}}$$

473 S', the total condensation surface, is made of the rod and the chamber sides, M is the molar 474 mass of the evaporated species and  $\alpha$  is the condensation coefficient (or sticking coefficient). 475 For a small slice of length dz, the total condensation surface is S'=  $2\pi$ (r+r') dz.

476 Considering the vapor phase as an ideal gas, one can write:  $\rho_i(z) = \frac{P_i(z)}{RT(z)}$ 

477 The flux conservation equation thus becomes:

478 
$$\rho_i(z)Sv = S' \frac{\alpha \left(P_i(z) - P_{i,sat}(z)\right)}{\sqrt{2\pi MRT(z)}} + \rho_i(z+dz)Sv$$

479 This equation can be further transformed as follows:

480 
$$\left(\frac{P_i(z)}{RT(z)} - \frac{P_i(z+dz)}{RT(z+dz)}\right)Sv = \frac{S'\alpha\left(P_i(z) - P_{i,sat}(z)\right)}{\sqrt{2\pi RT(z)}}$$

481 
$$\frac{d\left(\frac{P_i}{T}\right)}{dz}dz = \frac{2\pi(r+r')dz \ \alpha\left(P_i(z) - P_{i,sat}(z+dz)\right)}{Sv} \sqrt{\frac{R}{2\pi MT(z)}}$$

482 
$$\frac{T(z)\frac{dP_i}{dz} - P_i\frac{dT}{dz}}{T^2(z)} = \frac{2\pi(r+r')\alpha\left(P_i(z) - P_{i,sat}(z)\right)}{Sv}\sqrt{\frac{R}{2\pi MT(z)}}$$

483 The final equation is then written and can be integrated numerically:

484 
$$\frac{dP_i}{dz} = \frac{2\pi (r+r')\alpha \left(P_i(z) - P_{i,sat}(z)\right)}{Sv} \sqrt{\frac{RT(z)}{2\pi M}} + \frac{P_i}{T(z)} \frac{dT}{dz} \qquad (2)$$

The first term of the right-hand side of the equation corresponds to the condensation flux and the second term is the pressure variation linked to the temperature gradient dT/dz, which was computed with the calculated temperature from the Fluent software. The saturation pressures for the different species in the gas phase were evaluated with the equilibrium values calculated with FactSage.

490

491 Equation (2) was integrated numerically to calculate  $P_i(z)$  based on the computed thermal 492 gradient and the vapor pressure  $P_{i,sat}$  that was calculated in the thermodynamic simulations 493 described above. For the sake of simplicity, we focused on elements with a reduced number of species in the vapor phase and which condense in a reduced number of phases, such as  $Fe_{(g)}$ 494 which condenses in the FeSi molten alloy. We also applied this model to Si(g) in the upper part 495 496 of the rod, as it condenses in the FeSi alloy as well, and  $Ca_{(g)}$  which condenses only in the oxide melt and in the oldhamite when T > 1600 K. The fraction of element *i* that effectively condensed 497 relative to the maximum amount that would have condensed, had we reached chemical 498 499 equilibrium, can be expressed as:

500 
$$f_{i} = \frac{n_{i}(T) - n_{i}(T + \Delta T)}{n_{i}(T) - \overline{n_{i,sat}}(T)}$$
(3)

where  $n_i(T)$  is the number of moles of *i* at temperature T and  $\overline{n_{i,sat}}(T)$  is the mean number of moles of *i* in equilibrium over the [T, T+ $\Delta$ T] interval with  $\Delta$ T = 50 K. Our thermodynamic calculations show that  $\overline{n_{i,sat}}(T) \ll n_i(T)$  at all temperatures in the experiment. Thus, the value of  $f_i$  can be approximated with:

506 
$$f_i \approx \frac{n_i(T) - n_i(T + \Delta T)}{n_i(T)}$$
(4)

The  $f_i$  value depends implicitly on the oversaturation of a species  $P_i/P_{(sat,i)}$  in the vapor and on 507 the condensation coefficient  $\alpha$ , which is a critical parameter to compute the value of  $f_i$ . The 508 condensation coefficient of iron was determined experimentally<sup>19</sup> at pressures lower than those 509 of our experiment and reported a value of 1 for oversaturation greater than 10. However, it has 510 511 been shown for other elements that the condensation coefficient decreases with pressure. The 512 measured chemical pattern of Fe (Fig. 4) is best fit by an f value of 6 to 8%, corresponding to an  $\alpha$  value of 0.4, which is within the range of condensation coefficients reported in ref.<sup>19</sup> For 513 calcium, a value of 0.2 for  $\alpha_{Ca}$  yielded the best fit, which is consistent with published data for 514 Ca evaporation<sup>61,62</sup> and with the mean value for divalent cation evaporation from silicate 515 melt<sup>63,64</sup>, especially in H-rich environment<sup>61</sup>. The  $\alpha_{Si}$  is poorly constrained from previous work 516 and its value was adjusted to 0.45. The values of the condensation factors f are reported in Table 517 S3 for the various types of condensates. A best fit was obtained with a constant condensation 518 519 fraction for all chemical species of a given phase. For all the elements, we found  $f_i$  values well 520 below one (see Extended Data Table 3), expressing the kinetic limitation in the condensation 521 rate due to the effect of gas flow and kinetic barrier represented by the condensation coefficient 522  $\alpha_i$ .

523

524 As shown in Extended Data Fig. 4, the condensation fraction f of Fe assessed with this model 525 shows a moderate increase from 6% at 2280 K to 8.2% at 1377 K with a mean value of 7%. The computed  $f_{Si}$  value shows the same pattern with temperature, with a mean value equal to 526 527 8.3% in the region in which Si condenses in the FeSi metal. For Ca, two regime stages of 528 condensation were considered: the first region where Ca only condenses only in the oxide melt 529 (T>1750 K) and second a region where Ca condenses both in the silicate melt and in the sulfide phase (T<1750 K). This leads to a concave curve with an increase of  $f_{Ca}$  from 1.5% at 1950 K 530 531 to 2.6% at 1618 K. Finally, the f factor of the different phases used for the thermodynamic 532 calculations are presented in Extended Data Table 3.

533 534

# **D.** Optical properties and condensation distances

535 The condensation of dust in the outflow of C-rich stars should to some extent be controlled by the thermodynamics of solid phase formation with a vapor composition at a high C/O ratio. 536 However, it has been shown in several studies<sup>65</sup> that the radiative properties of the condensed 537 538 dust play a critical role on top of its thermodynamic properties. The photons radiated by the star 539 will interact with the dust: a fraction of the light will be reflected while another fraction will be 540 absorbed, and the last fraction is transmitted. If the fraction of absorbed light is significant, it 541 will contribute to heating the dust to a temperature that could be higher than the temperature of 542 the ambient gas. In turn, this higher temperature could delay dust condensation. This effect will contribute to increasing the oversaturation of this phase. As the gas and dust flow away from 543 544 the star, the temperature and pressure will decrease such that at some point the predicted dust temperature shall correspond to its condensation temperature<sup>39,40</sup>. At this radial distance the 545 546 dust can condensate.

547

In detail, the fraction of absorbed light depends critically on the optical parameters of the dust, 548 and it has been shown in numerous studies that this depends on the Fe content among other 549 parameters. When the Fe content increases, the absorption coefficient increases especially in 550 551 the frequency domain where radiations are efficient at heating the material. In the case of small dust particles ( $\mu$ m scale), the parameter used to describe light absorption is  $Q_{abs}$  and by plotting 552  $Q_{abs}/a$ , where a is the diameter of the grain, as a function of photon energy, one can define a 553 parameter p that is then used to calculate a dust temperature  $T_d^{40}$ . The parameter p has been 554 calculated for several oxides and for amorphous carbon but not for the mineral phases that are 555 predicted to condense in high C/O environment. Based on published optical properties of FeSi, 556

557 TiC, AlN, SiC and graphite we have calculated the value of parameter p to be able to predict 558 the corresponding dust temperature.

559 Ref<sup>40</sup> has shown that the distance of condensation of a particular mineral species can be 560 calculated based on the value of the parameter p and the temperature at the surface of the star 561  $T^*$ :

$$T_d(r) = T^* \left(\frac{2r}{R^*}\right)^{-\frac{2}{4+p}}$$
(1)

562 563

567

571 572

573

564 If the condensation temperature  $T_c$  is known independently from thermodynamic calculations, 565 it is then possible to determine the radial distance for condensation of a particular mineral phase 566 by setting:

$$T_d(r) = T_c(r) \tag{2}$$

For the purpose of our calculations, we have used the T(r) and P(r) curves for a C-rich star given in ref<sup>14</sup> and the results of our condensation calculations are presented graphically in figure 6.

#### E. Calculations of parameter p for mineral phases condensed at high C/O ratio

574 The value of p was calculated for the following mineral phases based on published optical 575 properties: TiC, AlN, CaS and FeSi. In the case of FeSi, the value of  $\alpha_{abs}$  the absorption 576 coefficient was calculated using the following equation that depends on the band gap energy<sup>66</sup>:

577 
$$\alpha_{abs} = \frac{A}{\omega} \sqrt{\omega - E_g}$$

578 Where  $E_g$  is the band gap energy. In the case of FeSi which is a semi-conductor at low 579 temperature but a metal at high temperature, the dependence of  $E_g$  on temperature was taken 580 into account with the following equation<sup>67</sup>:

581 
$$E_g = E_{g0} - Sh\omega. \left( coth\left(\frac{h\omega}{2kT}\right) - 1 \right)$$

582 The value of k, the imaginary part of the refraction index, was then calculated as:

583 
$$k(\lambda, T) = \frac{\alpha_{abs}}{4\pi}$$

584 Then, the value of the reflectivity given in ref<sup>68</sup> was used to calculate the value of the real part 585 of the complex refraction index (n+ik) using the following equation:

586 
$$R = \frac{(n-1)^2 + k}{(n+1)^2 + k}$$

587 Finally, the absorption parameter  $Q_{abs}$  was calculated using the following expression<sup>69</sup>:

588 
$$Q_{abs} = 4 \frac{2\pi a}{\lambda} \frac{6nk}{(n^2 - k^2 + 2)^2 + 4n^2k^2}$$

In the case of CaS, the experimental studies reported values of the real and imaginary dielectric function as a function of energy<sup>70</sup>. These parameters are directly related to the real  $\varepsilon_1$  and imaginary  $\varepsilon_2$  part of the complex refraction index:

592 593  $\varepsilon_1 = n(\omega)^2 - k(\omega)^2$  $\varepsilon_2 = 2n(\omega)k(\omega)$ 

594 From these equations, one can calculate:

595 
$$k(\omega) = \sqrt{\frac{\sqrt{\varepsilon_1^2 + \varepsilon_2^2} - \varepsilon_1}{2}}$$

596 
$$n(\omega) = \sqrt{\frac{\sqrt{\varepsilon_1^2 + \varepsilon_2^2} + \varepsilon_1}{2}}$$

The equation given above is then used to calculate  $Q_{abs}$ . In the case of TiC, the absorbance was 597 directly determined by ab initio calculation<sup>71</sup>. For AIN, the value of  $Q_{abs}$  is a function of 598 frequency that was directly calculated from the equation based on n and k, as given in  $ref^{72}$ . 599

#### 600 601

#### F. Modeling the timescale of grain growth relative to the timescale of outflows

The time necessary to grow a dust grain starting from seed such as a more refractory grain 602 highly depends on the local density of the constitutive atoms of the considered dust. If one takes 603 the example of FeSi (Extended Data Fig. 5), then the growth rate will be limited by the rate at 604 which individual atoms hit the grain surface. In the case of dust condensation in stellar outflow, 605 we calculated the maximum rate of condensation by neglecting the rate of evaporation. In all 606 607 cases, our calculated rates should be greater than the actual rates. As detailed in ref<sup>34,41</sup>, the condensation rate of FeSi is limited by the rate of Si abundance since it is lower than that of Fe. 608 609 One can thus write the timescale of grain growth as:

610 
$$\frac{da}{dt} = \frac{V_0 \alpha (P_{Si} - P_{Si,sat})}{\sqrt{2\pi m_{Si} RT}}$$

Where  $V_0$  is the average volume occupied by a FeSi unit. 611

This relation can be further modified following the study of ref<sup>41</sup> by including the relationship 612 between the local pressure, the density of the gas and the Si abundance: 613

614 
$$P_{Si} = \frac{n_{Si}}{n_H} \frac{2n_{H_2}RT}{V} = \varepsilon_{Si}\rho RT$$

In order to calculate the growth timescale as a function of distance from the star, one can use 615 the equation given in  $ref^{34}$ : 616

617 
$$\tau_{growth} = \frac{a}{\frac{da}{dt}} = \frac{a\sqrt{2\pi m_{si}RT}}{V_0 \alpha P_{si}} = \frac{a\sqrt{2\pi m_{si}RT}}{V_0 \alpha \varepsilon_{si} \rho RT} = \frac{2a}{V_0 \alpha \varepsilon_{si} \rho(r)} \sqrt{\frac{2\pi m_{si}RT}{RT}}$$

One can also calculate the timescale associated with the stellar outflow. According to ref<sup>47</sup> this 618 619 parameter can be evaluated with the following equation:

$$\tau_{outflow} = \frac{2}{\Gamma - 1} r \frac{v}{v_{es}^2}$$

Where  $\Gamma$  is defined as the ratio of radiative acceleration to gravitational attraction ref<sup>73</sup>: 621

$$\Gamma = \frac{f_K L_*}{4\pi c G M}$$

This parameter is increasing outwards and is above 1, ranging between 2 and  $5^{38}$  and  $v_{esc}$  is the 623 escape velocity: 624

$$v_{esc} = \sqrt{\frac{2GM^*}{4R^*}}$$

The velocity of the outflow at an infinite distance can be calculated with the following 626 equation<sup>38</sup>: 627

628 
$$v = \sqrt{\frac{2GM^*}{r_0}(\Gamma - 1) + v_0^2}$$

Where  $r_0$  represents the radius where the acceleration starts (the dust condensation radius). 629 According to ref<sup>38</sup> the initial velocity at  $R^*$  can be approximated as the sound velocity<sup>74</sup> that 630 can be defined as: 631

632 
$$v_0 = \sqrt{\frac{C_p^{H_2} RT}{M_{H_2} (C_p^{H_2} - R)}}$$

633 The value of Cp for H<sub>2</sub> was taken from the NIST compilation by ref<sup>75</sup>: 634

635 
$$C_p^{H_2} = 18.563083 + 12.257357 \times T - 2.859786 \times T^2 + 0.268238 \times T^3 + \frac{1.977990}{T^2}$$

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#### Author information

G.L. designed the research and performed the experiments with L.F., V-J.R., C.G. and F.C. G.L, C.G, V. G. and P. V. determined the mineralogy of the condensates. M.M., B.B. and G.L. reduced the data and did the thermodynamic calculations and models. G.L. M.M. and B.B. wrote the manuscript under the supervision of E.L for AGB implications.

#### Data availability

All data needed to evaluate the conclusions in the paper are present in the paper and in the Supplementary Information. Additional data related to this paper may be requested from the authors.

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# Extended data

# **Extended Data Table 1 Chemical Composition:**

Chemical compositions in wt % of the chondrite starting material and of the bulk composition of sampled condensates shown in Extended Data Fig. 1.

		Chemistry	Si	Al	Fe	Mn	Mg	Ca	Na	к	Ti	Р	0	С	S
		NWA 869 chondrite bulk composition	19.28	1.20	19.01	0.28	15.24	1.54	0.69	0.10	0.07	0.11	43.28	0.12	2.38
		Det. Limit	0.02	0.02	0.01	0.01	0.02	0.02	0.01	0.02	0.01	0.04			
Calculated average	# sample from	Depth of													
temperature*	top of T-rod	sampling													
in K		in mm													
2308	#14	0-50	24.95	0.16	42.81	0.02	0.03	0.09	< d.l.	< d.l.	0.14	< d.l.		26.28 ^	0.04
2142	#13	50-100	26.59	0.24	44.40	0.02	< d.l.	0.07	< d.l.	< d.l.	0.14	< d.l.		23.55 ^	0.04
2042	#12	100-150	31.61	0.79	57.07	0.12	< d.l.	0.06	< d.l.	< d.l.	0.25	< d.l.		2.59 ^	0.02
1949	#11	150-200	14.43	1.99	30.40	0.11	0.33	1.44	< d.l.	< d.l.	0.07	0.11		43.59 ^	0.43
1833	#10	200-250	13.70	1.24	18.18	0.13	8.46	1.68	< d.l.	< d.l.	0.04	0.13		38.37 ^	0.04
1741	#9	250-300	23.37	1.42	20.31	0.30	18.92	1.86	< d.l.	< d.l.	0.04	0.21		0.40	0.33
1685	#8	300-350	22.67	1.10	14.62	0.36	22.27	1.49	< d.l.	< d.l.	0.03	0.21		1.34	3.42
1615	#7	350-400	18.74	0.84	11.01	0.34	22.76	1.16	< d.l.	< d.l.	0.02	0.18		6.59	3.50
1556	#6	400-450	15.97	0.72	8.82	0.35	22.12	1.00	< d.l.	< d.l.	0.02	0.16		8.78	3.06
1502	#5	450-500	15.38	0.70	8.09	0.39	22.05	0.98	< d.l.	< d.l.	0.02	0.15		10.06	3.08
1453	#4	500-550	15.64	0.70	7.76	0.45	22.57	0.99	< d.l.	< d.l.	0.02	0.15		10.74	3.11
1395	#3	550-600	15.88	0.71	7.74	0.51	24.32	1.01	0.02	< d.l.	0.02	0.14		10.24	3.08
1389	#2	600-650	16.36	0.73	7.87	0.52	24.81	1.05	0.03	< d.l.	0.02	0.15		9.08	3.35
1365	#1	650-700	-	-	-	-	-	-	-	-	-	-		8.23	3.08

 $\label{eq:comparative} * Temperature\ calculated\ using\ the\ ANSYS\ Fluent\ computational\ fluid\ dynamics\ software\ (see\ methods).$ 

^ Carbon contamination of samples stuck to the carbon T-rod during sampling

# Extended Data Table 2 Mineralogy:

Mineralogy of the condensates sampled on the T-shape graphite rod holder as determined by Scanning Electron Microscopy and X-Ray diffraction.

		Mineralogy	с	FeSi	SiC	TiC	Al4SiC4	CaS	MgS	AIN	TiN	Si-glass	MgAl2O4	Mg2SiO4	MgO	MgSiO <sub>3</sub>	Notes
Calculated average temperature* in K 2308 2142 2042	# sample from top of T-rod #14 #13 #12	Depth of sampling in mm 0-50 50-100 100-150	-	↓ ↓ ↓	√-6Н √-6Н √-6Н	√	√					√ (Al-rich)					
1949 1833	#11 #10 #9	150-200 200-250 250-300	-		√-6H √-6H					1	$\checkmark$	√ √ √	$\checkmark$	1			
1685 1615	#5 #8 #7	300-350 350-400	***	× 				~	√			V V V		√ √	4	(√)	
1556 1502 1453 1395	#6 #5 #4 #3	400-450 450-500 500-550 550-600	* * *	* * *								↓ ↓ ↓		* * *	×	\ \ \	
1389 1365	#2 #1	600-650 650-700	~ ~	√ √								√ √		√ √	✓ √	$\checkmark$	

**Extended Data Table 3 Bulk Chemistry**: Bulk composition of the starting material and values of condensation factors used in the modeling. The composition of the experiment corresponds to a C/O of 0.93 and calculated considering that 600 g of NWA869 ordinary chondrite were introduced with  $4 \text{ Nm}^3$  of Ar and  $2\text{Nm}^3$  of H<sub>2</sub>.

Element	mass (g)	Phase	f %			
Fe	111.5	FeSi	8			
Mn	1.617					
Ti	0.390	SiC	3			
Ca	9.045	AIN	5			
Κ	0.575		5			
Ar	7132	CaS	2			
S	13.96					
Р	0.648	Silicate melt	4			
Si	113.1	Aluminates	2			
Al	7.061		-			
Mg	89.41	Olivine	8			
Na	4.061	MaO	5			
0	248.63	IvigO	5			
Ν	150	TiC	5			
С	173.58	Cronhite	o			
Н	179.94	Graphile	0			

Phase	р	Reference
Graphite	2.0	76
SiC	0.3	77
TiC	0.8	71
FeSi	2.2	68
AlN	-0.35	72
CaS	0.82	70
Mg <sub>2</sub> SiO <sub>4</sub>	-0.9	78
MgSiO <sub>3</sub>	-0.5	39

Extended Data Table 4 Optical Parameter: Optical parameter p of various mineral phases.

# Extended Data Fig. 1 2D Thermal Regime.

The 2D thermal regime (a) inside the chamber as well as the trajectory (b) and velocity of fictive particles (c) have been obtained using the ANSYS Fluent computational fluid dynamics software (see Methods).



#### Extended Data Fig. 2 Samples.

Photograph of the graphite rod sample holder after the experiment that collected the condensates. As shown, 14 segments of 5 cm each have been sampled for bulk chemical analysis. CS1-CS6 correspond to sampling for detailed mineralogy depicted in Extended Data Fig. 3. See the chemistry and mineralogy of the condensates in Extended Data Table 1 and Table 2, respectively.



#### Extended Data Fig. 3 Further Mineralogy.

Further mineralogical and chemical characterization of condensed phases sampled on the graphite T-rod sample holder with location indicated on Extended Data Fig. 3. All images and analyses by scanning electron microscopy (SEM) and energy dispersive X-ray (EDX) analysis, unless specified.

CS1 and CS2 (#14)



Notice in the highest temperature condensates the abundance of crystallized hexagonal SiC bathed in an iron silicide liquid, now crystallized in a mixture of naquite (FeSi), hapkeite (Fe<sub>2</sub>Si) and possibly xifengite (Fe<sub>5</sub>Si<sub>3</sub>).

CS1 and CS2 (#14)



Titatium (TiC) and aluminum  $(Al_4SiC_4)$  carbides are also present in close association with the iron silicides.

CS4 (#11)



Intermediate condensates contain aluminum (AlN) and titanium (TiN) nitrides. Notice the remarkable 1-D whisker of AlN formed by a FeSi catalyst-assisted VLS (vapor-liquid-solid) growth process. All 3 first images by SEM, excepted the bottom one right which corresponds to transmission electron microscopy (*TEM*) analysis and the obtained diffraction pattern of AlN. CS4 (#11)



Between 2000-1900 K  $Mg_2SiO_4$  starts to crystallize from the melt (now glassy and enriched in MgO and SiO<sub>2</sub>) in close association with FeSi (black beads). Panchromatic images obtained by high resolution cathodoluminescence.

CS5 (#8)



Below a temperature range of 1850-1800 K, carbides are no longer detectable, and olivine, FeSi and sulfides (Ca,Mg)S and graphite are frequently observed..

CS5 (#8)



Below 1800K, the average size of condensed phases becomes micron-sized. Remarkable wiskers of enstatite are present.

CS6 (#2)



The lowest condensates on the T-shape graphite holder becomes very fine grains more difficult to identify by SEM. A bulk analysis by EDX of a few  $\mu m^2$  of condensates reveals a composition similar to that obtained by ICP-MS on a larger sample (Fig.4).



In these range of temperature circa 1400 K, the mineralogy is dominated by magnesian silicates (olivine and pyroxene), sulfides, oxides (MgO), with graphite and silicides (Fe/Si $\approx$  2).

#### Extended Data Fig. 4 Kinetic Model.

Schematic view of the kinetic model. a) The internal cylinder is the graphite rod. The total surface of evaporation corresponds to the graphite rod and the chamber's sides. b) Condensation factor of Ca (red) and Fe (blue). For temperatures higher than 1750 K, Ca partitions only in the oxide melt, then in oxide melt and CaS



#### Extended Data Fig. 5 FeSi Growth Timescale.

Calculated timescales of FeSi growth (red) and stellar outflow (blue) as a function of the distance of the star (normalized to star radius R\*). Red curves are for Si growth for grains with a size labeled on the curve. Assuming a larger grain diameter would lead to longer timescales. Sticking coefficient of Si<sub>(g)</sub> is taken from ref.<sup>79</sup>.

