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1 **Metal segregation in planetesimals:**
2 **constraints from experimentally determined interfacial energies**

3

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8 Keywords: Metal-silicate differentiation, interfacial energies, high temperature experiments,
9 planetesimals, primitive achondrites

10

11 **Abstract**

12 High temperature experiments have been performed to constrain interfacial energies in a
13 three-phase system (metal – forsterite - silicate melt) representative of partially
14 differentiated planetesimals accreted early in the solar system history, with the aim of
15 providing new insights into the factors affecting the interconnection threshold of metal-rich
16 phases. Experiments were run under controlled oxygen fugacity ($\Delta_{\text{Ni-NiO}} = -3$) at 1440°C,
17 typically for 24h. Quantification of the true dihedral angles requires a resolution of at least
18 30 nm per pixel in order to reveal small-angle wedges of silicate melt at crystal interfaces.
19 At this level of resolution, dihedral angle distributions of silicate melt and olivine appear
20 asymmetric, an observation interpreted in terms of anisotropy of olivine crystals. Based upon
21 the theoretical relation between dihedral angles and interfacial energies in a three-phase
22 system, the relative magnitudes of interfacial energies have been determined to be: $\gamma_{\text{Melt-OI}} <$

23 $\gamma_{\text{Melt-Ni}} < \gamma_{\text{Ol-Ni}}$. This order differs from that obtained with experiments using an iron sulfide
24 liquid close to the Fe-FeS eutectic for which $\gamma_{\text{Melt-Sulfide}} < \gamma_{\text{Melt-Ol}} < \gamma_{\text{Ol-Sulfide}}$, implying a lower
25 interconnection threshold for sulfur-rich melts than for pure metallic phases. This
26 dependence of the interconnection threshold on the sulfur content will affect the drainage
27 of metallic phases during melting of small bodies. Assuming a continuous extraction of
28 silicate melt, evolution of the metal volume fraction has been modeled. Several sulfur-rich
29 melts extraction events are possible over a range of temperatures relevant with
30 thermometric data on primitive achondrites (1200-1400°C and 25% of silicate melt
31 extracted). These successive events provide novel insight into the variability of sulfur content
32 in primitive achondrites, which are either representative of a region that experienced sulfide
33 extraction or from a region that accumulated sulfide melt from overlying parts of the parent
34 body.

35 **1 Introduction**

36 Metal-silicate differentiation in early-accreted bodies of the solar system involves
37 processes that are still poorly understood. Metal connectivity is a key factor because
38 efficient phase separation requires an interconnected network of the metal-rich phase. In
39 other words, the interconnection threshold (i.e. the volumetric phase fraction at which the
40 phase forms a continuous network) has to be reached. This threshold is often assumed to
41 have a very low value (3 to 6 vol%, e.g. Šrámek et al., 2012). However, the meteoritic record
42 shows evidence of incomplete metal segregation. For example, primitive achondrites (e.g.

43 Acapulcoites and Lodranites) are partially differentiated natural samples that exhibit signs of
44 silicate partial melting, up to 25 vol% (e.g. Bild and Wasson, 1976; McCoy et al., 1997, 1996;
45 Palme et al., 1981; Papike et al., 1995; Zipfel et al., 1995) and have FeNi alloy contents up to
46 ~20 vol% and troilite contents of ~3 vol% (Bild and Wasson, 1976; Palme et al., 1981).
47 Under these conditions, the metal content is larger than commonly assumed interconnection
48 thresholds and these bodies should be fully differentiated, which they are clearly not. To
49 rationalize this observation, either metal-rich phases did not readily form a connected
50 network and/or the kinetics of metal segregation were slow compared to cooling time-
51 scales. While this latter possibility cannot be excluded for the case of primitive achondrites -
52 because the parent bodies were planetesimals of modest radius (Neumann et al., 2018), the
53 gravitational driving force for segregation was low - the question of relevant values of
54 segregation threshold must be treated first.

55 In general terms, the interconnection threshold is a function of interfacial energies that
56 drive the geometry of local phase boundaries to minimize the energy of the system (Bulau
57 et al., 1979; Jurewicz and Watson, 1985; Smith, 1964). Thus, the system will evolve to shorten
58 high-energy interfaces at the expense of lower energy geometries. Physical expression of
59 these effects corresponds to the set of dihedral angles that form at phase triple junctions
60 (i.e. where three phases meet). In a two-phase system (e.g. melt-mineral), the dihedral angle
61 θ , corresponding to the angle of the melt between two solid grains, is described by
62 equation (1) (Smith, 1964).

63
$$2 \cos \frac{\theta}{2} = \frac{\gamma_{SS}}{\gamma_{SL}} \quad (1)$$

64 where γ_{SS} is the solid-solid interfacial energy and γ_{SL} the solid-liquid interfacial energy (in
65 J.m⁻²).

66 Measuring dihedral angles is thus a way to quantify the ratios of interfacial energies and
67 to constrain variations in interconnection threshold. Two methods are commonly used to
68 measure dihedral angles: the first one is the sessile drop technique using in-situ X-Ray
69 imaging a droplet on a solid substrate (e.g. Humenik Jr. and Kingery, 1954; Jung et al.,
70 2010). The second one consists in a more statistical approach with repeated measurements
71 on 2D scanning electron images. In this work we use the second method was preferred as it
72 allows more accurate imaging of the triple junctions. In the case of a two-phase system,
73 such as a metallic melt in an olivine matrix, the relation between interconnection threshold
74 and dihedral angle is well established (e.g. Laporte and Provost, 2000). For instance, an iron-
75 dominated metallic liquid typically has a high dihedral angle of 110-130° in an olivine matrix
76 (Gaetani and Grove, 1999; Shannon and Agee, 1996), corresponding to an interconnection
77 threshold of roughly 10 to 15 vol% (Laporte and Provost, 2000). Adding light elements (C,
78 O, S) to this metallic melt decreases its dihedral angle, and therefore its interconnection
79 threshold (Ballhaus and Ellis, 1996; Gaetani and Grove, 1999; Holzheid et al., 2000; Minarik et
80 al., 1996). For compositions in the vicinity of the Fe-FeS eutectic, dihedral angles span a
81 range from 60° up to 90°, yielding a broad range of interconnection thresholds from 0 to 7
82 vol%, as estimated from Laporte and Provost (2000). However, as mentioned above,

83 segregation is not efficient in such systems, core formation being too slow to match with
84 the meteoritic record (Bagdassarov et al., 2009). Adding a silicate melt makes the
85 experimental system more realistic and tends to promote more efficient segregation
86 (Bagdassarov et al., 2009). However, equilibrium geometries of such three-phase systems
87 (crystalline silicate + silicate melt + pure metal or sulfide) have received little attention. To
88 our knowledge, data are only available for a Fe-FeS eutectic composition (Holzheid et al.,
89 2000) in a partially molten system of San Carlos olivine and basalt.

90 During the evolution of a planetesimal, composition of the metallo-sulfide phase ranges
91 from the eutectic composition, representing the first metal-rich liquids produced, to pure
92 metal (FeNi). The present study focuses on the measurement of dihedral angles and
93 equilibrium geometries in that latter end-member. Constraints are obtained using high
94 temperature experiments in a one-atmosphere furnace, a pressure relevant given the low-
95 pressure environment in early-accreted planetesimals. Equilibrium textures in the
96 experimentally produced samples containing partially molten silicate and pure metal are
97 then used to construct a global metal-silicate differentiation model that can help to
98 rationalize the observed mineralogy of primitive achondrites.

99 **2 Experimental setup**

100 Experiments were performed in a high temperature, 1 bar, controlled-atmosphere vertical
101 furnace, using a three-phase system. The experiments allow to determine the equilibrium

102 geometries between forsterite, silicate melt and nickel metal with accurate quantification of
103 the dihedral angles and associated interfacial energies.

104 **2.1 Synthesis of starting material**

105 Samples are mixtures of crystalline forsterite (Fo), silicate melt (Liq) and nickel metal (Ni).
106 The starting material was prepared in two steps.

107 First, a silicate glass was synthesized, with a composition in equilibrium with pure
108 forsterite at 1430°C as determined from the Anorthite-Diopside-Forsterite ternary phase
109 diagram in the CMAS (CaO – MgO – Al₂O₃ – SiO₂) system (Morse, 1980), a good analogue
110 of iron-free silicate melts in meteorites. Thus, the proportion of silicate melt (of fixed
111 composition) can be varied at a single temperature. Oxide powders (SiO₂ 99.8% – Alfa Aesar,
112 Al₂O₃ 99.999% extra pure – Acros) and carbonates (CaCO₃ 99+% - Acros,
113 4(MgCO₃).Mg(OH)_{2.5}(H₂O) - Acros) were mixed in the appropriate proportions to produce
114 the desired composition of silicate melt (Table 1). This mixture was finely ground, placed in
115 a platinum crucible and held for two hours at 1100°C to allow complete dehydration and
116 decarbonation, then heated to 1500°C for two hours for melting. The material was
117 quenched, ground and put back in the furnace at 1500°C for two additional hours to ensure
118 chemical homogeneity. After quenching, glass composition was checked using Electron
119 Micro Probe Analysis (EMPA, Table 1). Finally, the glass was finely crushed before mixing
120 with the other components.

121 Although metal is dominated by iron in natural samples, nickel has been chosen in these
122 experiments in light of its lower melting point and its stability at higher oxygen fugacity
123 (fO_2). This choice allowed performing experiments (at conditions of temperature and fO_2
124 that are easily obtained and controlled) in which the metallic subsystem does not interact
125 significantly with the silicates, i.e. the diffusion of Ni in forsterite and silicate melt is low (see
126 section 2.2).

127 In the second stage of sample synthesis, the glass was mixed with commercial powders
128 of forsterite (Neyco CERAC, Inc., 325 mesh, 99% pure; grain size 0-40 μm) and nickel
129 (Goodfellow, 99.8% pure, grain size 1-70 μm), with fixed metal content but varying forsterite
130 / glass ratios in order to investigate the effect of the relative abundance of these two
131 phases (Fo : Liq : Ni expressed in vol%): 20:75:5, 47.5:47.5:5, 62.5:32.5:5, 75:20:5. The
132 obtained mixtures were then sintered to reduce porosity as the gas trapped between grains
133 may act as a fourth phase with its own interfacial energy. Spark Plasma Sintering (SPS) was
134 used at the PNF² platform (University Paul Sabatier, Toulouse, France), in an argon vacuum,
135 at 100 MPa and 1200°C for 5 minutes, as described by Guignard et al. (2011). The 20:75:5
136 mixture could not be sintered by SPS due to devitrification around 900°C that caused the
137 graphite mold to break due to the large change in volume of the sample. In this case, the
138 wire loop technique was employed using a platinum wire ($\varnothing = 0.2$ mm, 99.9% pure –
139 Johnson Matthey) on which ~30 mg of sample was placed and put for 30 seconds at
140 1450°C to form a droplet held on the loop by surface tension.

141 **2.2 Annealing experiments**

142 Experiments were performed in the vertical furnace at a fixed temperature of 1440°C,
143 ~15°C below the melting point of nickel. Temperature was measured ($\pm 1^\circ\text{C}$) using an S-
144 type thermocouple (PtRh_{10%}/Pt). The oxygen fugacity was fixed by mixing appropriate
145 proportions of CO/CO₂ yielding a $\log(f\text{O}_2)$ of -8.46 atm. (i.e. $\Delta_{\text{Ni-NiO}} = -3$). Such reducing
146 conditions allow nickel grains to remain in their metallic state and limit diffusion and
147 dissolution into silicates. Annealing experiments were run for 24 hours for all samples and
148 an additional experiment of 10 days was performed on the 75:20:5 mixture to check
149 attainment of textural equilibrium.

150 **2.3 Analytical techniques**

151 After experiments, samples were mounted in epoxy resin and polished down to 0.3
152 micron using Al₂O₃ grit for 2 hours. FEG-SEM (Field Emission Gun Scanning Electron
153 Microscope) and EPMA (Electron Probe Micro Analysis) were then performed.

154 Textures and dihedral angles were analyzed with the image processing software Fiji
155 (Eliceiri et al., 2012; Schindelin et al., 2012) from digital images acquired by backscattered
156 electron detection, secondary electron detection, or stacking both kinds of images, on a
157 JEOL JSM-7800F Prime FEG-SEM at the Raimond Castaing center in Toulouse (France).
158 Images were acquired using a 10 keV accelerating voltage and a 10 nA beam current. There
159 are two levels of organization of triple junctions depending on the scale of observation. At
160 macroscale, the minimization of interfacial energies has a weak effect on the geometry that

161 is principally controlled by crystallographic orientation. On the other hand, at microscale, at
162 the apex of the triple junctions, geometry is dictated by the minimization of interfacial
163 energies between the three phases. To quantify a potential shift from a macroscopic to a
164 microscopic behavior, various magnifications and thus resolutions were investigated: x250
165 (376 nm per pixel), x1000 (94 nm per pixel), x3000 (30 nm per pixel) and x10,000 (9.4 nm
166 per pixel).

167 Quantitative analyses of the chemical composition of the phases were performed using a
168 Cameca SXFive EPMA at the Raimond Castaing Center (Université Paul Sabatier, Toulouse,
169 France). The operating conditions were: accelerating voltage 15 keV; beam current 20 nA;
170 analysed surface of $\sim 2 \times 2 \mu\text{m}^2$. Periclase, corundum, wollastonite, platinum and NiO were
171 used as standards for Mg, Al, Ca and Si, Pt, and Ni respectively.

172 **3 Dihedral angle measurement**

173 **3.1 Chemical and textural equilibrium**

174 Chemical equilibrium was checked using EPMA on the two silicate phases (Table 1). The
175 composition of the glass is found to not perfectly match the equilibrium composition
176 expected at 1450°C. This difference can be explained by crystallization of forsterite (~ 4 wt%)
177 during sample quench. NiO was detected in forsterite and silicate glass, at levels of 2 wt%
178 and 0.6 wt%, respectively (Table 1). No concentration profile was observed in either of these
179 phases, which is in agreement with diffusion and partitioning coefficients previously

180 obtained at similar experimental conditions (Borisov, 2006; Dingwell et al., 1994; Matzen et
181 al., 2017; Petry et al., 2004), indicating chemical equilibrium.

182 Textural equilibrium was checked using SEM images (Figure 1). In the 20:75:5 and
183 47.5:47.5:5 samples, nickel grains are elongated in comparison with the other samples. A
184 remarkable feature of the 20:75:5 sample is that nickel is clustered at the center of the
185 sample and forms an interconnected network. Looking at textures at the grain scale, similar
186 features and olivine triple junctions at 120° are observed in all samples. There are no
187 differences in the textures or in the dihedral angle distributions (see 3.3) between 24 hour-
188 and 10 day-experiments. Hence, at triple junction scale, textural equilibrium is reached.

189 **3.2 True dihedral angle measurement**

190 Dihedral angles of silicate melt and forsterite were measured using the "angle tool" of
191 the Fiji distribution of ImageJ, while the dihedral angle of nickel was inferred from the two
192 others, considering that the sum of the three dihedral angles is 360° . In order to obtain
193 sufficient statistics, measurements were repeated on at least 100 different triple junctions.
194 These measured distributions are called apparent dihedral angle distributions, as they are
195 representative of a 2D slice in a 3D volume. For a single population of dihedral angles, these
196 apparent distributions can be fitted with a Gaussian function centered on the median value
197 (Jurewicz and Jurewicz, 1986), that corresponds to the true dihedral angle, i.e. the dihedral
198 angle in the 3D volume.

199 3.2.1 From macroscopic to microscopic behavior of triple junctions

200 Figures 2a, 2b and 2c display the true dihedral angle for each phase in each sample as a
201 function of pixel size. With increasing detail (i.e. decreasing pixel size), the dihedral angle of
202 the melt decreases from $\sim 70^\circ$ to $\sim 25^\circ$, and then reaches a steady value within uncertainty,
203 as shown by the measurements at a resolution of 9.4 nm per pixel. The dihedral angles of
204 forsterite and Ni exhibit the opposite trend upon increasing resolution: they increase from
205 125° to 160° for forsterite and from 160° to 175° for Ni, until a constant value is reached.
206 These observations indicate that micro-films and/or micro-channels of silicate melt are
207 present between forsterite and/or Ni grains, as previously suggested by Cmíral et al. (1998).
208 For low melt fractions, distribution of the melt can be anisotropic (Faul, 2000): some grain
209 boundaries are still dry whereas the others are wetted by silicate melt, and the width of
210 channels between grains may vary from 10 to 100 nm, making resolution a key parameter
211 to image melt distribution (Garapić et al., 2013). In the present study, close to triple
212 junctions, these structures represent hooks between forsterite and nickel grains that are
213 thinner and thinner toward the apex of the triple junction (Figure 1g and 1h). Typically, the
214 width of these structures at half-length of the hook is about 100 nm. To be accurately
215 measured, we considered that at least 9 pixels are needed, i.e., a resolution threshold of 30
216 nm per pixel corresponding to a x3000 magnification in our case. At lower resolution, the
217 continuous evolution of dihedral angle as a function of magnification indicates a continuous
218 evolution from the microscopic to the macroscopic energy balance.

219 It is also of note that at low resolution (376 and 94 nm per pixel), dihedral angles appear
220 to be a function of phase proportions. For instance, the 20:75:5 and the 47.5:47.5:5 samples
221 (i.e., where Ni grains are elongated and seem to form a network) show dihedral angles of
222 roughly 60° for the silicate melt and 170° for the nickel, whereas the 62.5:32.5:5 and the
223 75:20:5 samples (i.e. samples displaying isolated beads of nickel) exhibit angles that are 20°
224 higher for silicate melt and 15° lower for nickel (Figure 2a and 2c). At higher resolution this
225 difference is no longer apparent, highlighting the same microscopic behaviors but different
226 macroscopic ones.

227 Once all the small-scale details are unraveled, all samples display the same dihedral
228 angle distribution and the same values of true dihedral angles, regardless of the proportions
229 of each phase. In light of this result, the different datasets for a same magnification were
230 merged to provide better statistics on the dihedral angle distribution. In the remainder of
231 the text, the median values of these new merged distributions are considered as the true
232 dihedral angles (Figure 2, Table 2).

233 **3.2.2 Macroscale geometry: apparent isotropy**

234 Figures 2d and 2e show the merged dihedral angle distributions at 376 and 94 nm per
235 pixel and the associated Gaussian fits for each phase. These distributions are fitted with an
236 unimodal Gaussian curve, as suggested by Jurewicz and Jurewicz (1986). The quality of the
237 fit is better for the 376 nm per pixel distributions than for that at 94 nm per pixel, which is
238 due to a slight asymmetry of the latter and an intermediate behavior between macro- and

239 micro-scales. The good agreement between data and unimodal Gaussian curves suggests
240 that there is a single population of dihedral angles, so at this macroscale, olivine grains
241 appear isotropic, as commonly assumed in most previous studies of dihedral angles in this
242 type of system.

243 **3.2.3 Microscale geometry: anisotropy of forsterite grains**

244 At the microscale, i.e. close to the apex of the triple junctions (Figure 2f, 30 and 9.4 nm
245 per pixel), distributions for forsterite and silicate melt become clearly asymmetric, with the
246 emergence of a narrow peak and a shoulder towards lower values for forsterite, and higher
247 values for silicate melt. In this case, data cannot be fitted by a unimodal Gaussian function.
248 Similar features appear when angle distributions are mixed (Jurewicz and Jurewicz, 1986),
249 indicating a possible difference of interfacial energies along crystal faces. In this respect, it
250 has been shown that wetting behavior of olivine depends on crystal orientation (e.g. Jung
251 and Waff, 1998; Wanamaker and Kohlstedt, 1991). Hence, the asymmetry of the distributions
252 may be linked to different populations of dihedral angles reflecting the anisotropic behavior
253 of forsterite. On the other hand, dihedral angle distribution of nickel remains very
254 symmetric, which can be explained by its cubic crystalline system for which all crystal planes
255 are equivalent. Finally, due to its low viscosity, silicate melt accommodates the anisotropy of
256 forsterite and has thus different dihedral angle populations due to different olivine
257 orientations. Three principal populations of interfacial energies exist in forsterite crystals
258 (Jung and Waff, 1998), corresponding to Miller indices of (010), (110) and a family of minor

259 crystal planes ((021), (120), (001), (101) and (100)). Hence, three Gaussian functions were
260 fitted on both silicate melt and forsterite dihedral angle distributions (Figures 2f, 2g and 2h).
261 The agreement between measured and calculated distributions is characterized by a
262 correlation greater than 0.95 (inset in Figures 2g and 2h). The newly obtained median values
263 for each population are 15.2°, 35.5° and 52.9° for the silicate melt and 173.1°, 162.2° and
264 137.5° for the olivine, each pair of dihedral angles corresponding to a new subset called 1, 2
265 and 3 respectively (Table 3). Calculating the area under each Gaussian provides the
266 proportion of each population of interfacial energies, corresponding roughly to ~33 % for
267 each distribution. These values are in good agreement with the proportions of the
268 crystallographic planes given by Jung and Waff (1998).

269 **4 Interfacial energies and implications for metal segregation in** 270 **planetesimals**

271 **4.1 Three-phase system: theoretical approach**

272 In a three-phase system, the equilibrium geometry of a triple junction can be
273 represented as in Figure 3. When textural equilibrium is reached, the three different dihedral
274 angles θ_1 , θ_2 and θ_3 vary as a function of the interfacial energies γ_{12} , γ_{13} and γ_{23} according to
275 equation (2), assuming crystalline isotropy (Laporte and Provost, 2000; Smith, 1964).

$$276 \quad \frac{\gamma_{12}}{\sin(\theta_3)} = \frac{\gamma_{13}}{\sin(\theta_2)} = \frac{\gamma_{23}}{\sin(\theta_1)} \quad (2)$$

277 In detail, given that olivine crystals are anisotropic (see above), this equation can be used for
278 each pair of olivine – silicate melt dihedral angle population. Rearranging this equation
279 yields a set of three equations:

$$280 \quad \frac{\sin(\theta_1)}{\sin(\theta_2)} = \frac{\gamma_{23}}{\gamma_{13}} \quad \frac{\sin(\theta_2)}{\sin(\theta_3)} = \frac{\gamma_{13}}{\gamma_{12}} \quad \frac{\sin(\theta_1)}{\sin(\theta_3)} = \frac{\gamma_{23}}{\gamma_{12}} \quad (3)$$

281 Based on this equation, the ratio of two interfacial energies can be inferred by calculating
282 the ratios of sines of dihedral angles. From a theoretical point of view, it is thus possible
283 to illustrate how variations in phase geometry translate into variations in the ratio of
284 interfacial energies (Figure S1). Given that an individual dihedral angle cannot be smaller
285 than 0° and no larger than 180°, and that the sum of the three dihedral angles is
286 constrained to be 360°, the variation of interfacial energy ratio can be represented using a
287 ternary diagram. Due to the aforementioned conditions, only the inner triangle of the
288 ternary diagram is of interest (Figure 4a). From equation (3), equality between two interfacial
289 energies can be achieved when two dihedral angles are equal. Plotting these lines in the
290 ternary diagram defines six different energetic regimes. In each of these six domains, the
291 relative order of interfacial energies differs (Figure 4b). The central point of this diagram
292 represents the case where each phase has a dihedral angle of 120° (i.e. all interfacial
293 energies are equal).

294 While a given ratio of interfacial energies always leads to the same geometry at the scale
295 of an individual triple-junction, the larger-scale distribution of material will be affected by

296 phase proportions, and limited by the least abundant phase. Given that olivine is always the
297 major phase, the range of potential geometries is illustrated for the cases of nickel (Figure
298 4c) or silicate melt (Figure 4d) as the minor phase. For instance, we consider the case where
299 two phases have dihedral angles of 180° and the third one has a dihedral angle of 0° . As
300 displayed on Figure 4c and 4d, in this situation the third phase entirely wets the two others,
301 preventing contact between them. If the third phase is present in small proportion, it will
302 form a thin film along the interface between the two others. Alternatively, if one of the two
303 other phases is limited in proportion, this phase will form an isolated sphere into the third
304 phase (Figure 4c and 4d).

305 Figures 4c and 4d also represent a first order estimation of the interconnection threshold
306 – dihedral angle dependence. When a phase has a dihedral angle close to zero, it has a thin
307 lens shape, spreading along the interface of the two other phases. In this case, its
308 interconnection threshold is very low. Increasing the value of the angle leads to an
309 increasingly spherical shape and higher interconnection threshold.

310 **4.2 Interfacial energy ratios calculation**

311 Interfacial energies can theoretically be calculated from the knowledge of the dihedral
312 angles, if at least one interfacial energy is known independently. In this respect, the
313 interfacial energy between silicate melt and olivine measured by Cooper and Kohlstedt
314 (1982) seems a suitable reference point ($0.5 \pm 0.2 \text{ J.m}^{-2}$). However, this value assumes
315 isotropy of crystalline olivine, a situation that does not reflect reality as discussed above. To

316 our knowledge, no study reports different interfacial energies for different orientations of
 317 olivine crystals, so that it is preferable to stick to calculating ratios as follows:

$$318 \quad \frac{\gamma_{Melt-Ni}}{\gamma_{Melt-Fo}} = \frac{\sin(\theta_{Fo})}{\sin(\theta_{Ni})} \quad \frac{\gamma_{Fo-Ni}}{\gamma_{Melt-Fo}} = \frac{\sin(\theta_{Melt})}{\sin(\theta_{Ni})} \quad \frac{\gamma_{Melt-Ni}}{\gamma_{Fo-Ni}} = \frac{\sin(\theta_{Fo})}{\sin(\theta_{Melt})} \quad (4)$$

319 Results of these calculations are shown in Table 3 for the different crystal plane families
 320 (i.e. subsets) and for the complete dataset (referred to as "merged"). When the raw data are
 321 treated, the sum of the three dihedral angles is greater than 360° for each subset. Given
 322 that this result is impossible, angles have been recalculated by fixing the nickel dihedral
 323 angle to 176.1° (corresponding to the measured value), then normalizing the two remaining
 324 values such that the sum is 360°.

325 The three-phase system considered composed of silicate melt, forsterite and nickel plots
 326 in the $\gamma_{Melt-Fo} < \gamma_{Melt-Ni} < \gamma_{Fo-Ni}$ domain as shown in Figure 5 (using data from Table 2, column
 327 "Merged", and from Table 3). At low resolution, nickel grains seem to be almost equally in
 328 contact with the forsterite and the silicate melt, whereas at high resolution, they are more
 329 rounded and barely in contact with forsterite crystals. This behavior is illustrated through the
 330 variation of the interfacial energy ratios as a function of magnification. At low resolution
 331 (376 and 94 nm per pixel) both $\gamma_{Melt-Fo}/\gamma_{Melt-Ni}$ and $\gamma_{Melt-Fo}/\gamma_{Fo-Ni}$ ratios are almost equal, while
 332 at higher resolution (30 and 9.4 nm per pixel) these two energy ratios are lower and Ni has
 333 a higher interfacial energy than the rest of the system.

334 The energy ratios involving olivine are quite different depending on crystal orientation.
335 From subset 1 to subset 3, the $\gamma_{\text{Melt-Fo}}/\gamma_{\text{Melt-Ni}}$ and $\gamma_{\text{Melt-Fo}}/\gamma_{\text{Fo-Ni}}$ ratios decrease from 0.3 to 0.1,
336 and the $\gamma_{\text{Melt-Ni}}/\gamma_{\text{Fo-Ni}}$ ratio increases toward values close to 1, illustrating an evolution in the
337 geometry toward the $\gamma_{\text{Melt-Ni}} = \gamma_{\text{Fo-Ni}}$ isovalue line (Figure 5). The changes of equilibrium
338 geometries for the different subsets can be illustrated using Figure 4. For example, in case
339 of low metal fraction, the metal grain becomes less embedded in forsterite from subset 3 to
340 subset 1, making it easier to form an interconnected network in the latter case. This leads to
341 an interconnection threshold that varies locally, increasing from subset 1 to subset 3. As
342 interfacial energies are the lowest for subset 1, this geometry will most likely dominate the
343 overall distribution. However, the energetic effect is countered by samples composed of an
344 aggregate of randomly oriented crystals leading to metal grains in contact with various
345 olivine crystal planes, including crystalline planes of high energy. To reach a state of lower
346 energy, a dynamic process such as deformation would be required to re-orientate olivine
347 grains. Despite these differences, it is of note that all subsets of olivine orientation fall in the
348 same energetic regime where $\gamma_{\text{Melt-Fo}} < \gamma_{\text{Melt-Ni}} < \gamma_{\text{Fo-Ni}}$.

349 In order to infer the mobility of sulfur-rich and pure metallic melts in an olivine matrix,
350 the experimental results of Ballhaus and Ellis (1996) can be used. Calculating the interfacial
351 energy ratios from their measured dihedral angles gives $\gamma_{\text{Ol-Ol}}/\gamma_{\text{Ol-FeS}} = 1.463$ to 1.658 and $\gamma_{\text{Ol-}}$
352 $\text{Ol}/\gamma_{\text{Ol-Fe}} = 1.075$ to 1.104 . Taking these ratios at face-value (note that calculated values are
353 based on angles measured by Ballhaus and Ellis (1996) at a resolution that was not

354 specified) suggests that $\gamma_{\text{Ol-Ol}} \approx \gamma_{\text{Ol-Fe}} > \gamma_{\text{Ol-FeS}}$. In such a two-phase system, the olivine –
355 metal-rich phase interfacial energy is lower in the case of sulfide than of pure metal. Thus
356 sulfide melts appear to wet olivine grains more than pure metallic ones do, and sulfur-rich
357 melts thus have a lower interconnection threshold (~3 to 7 vol% from Laporte and Provost,
358 2000) than pure metallic melts in an olivine matrix (~12 vol% from Laporte and Provost
359 (2000), although the high dihedral angles determined in this study suggest a much higher
360 interconnection threshold on the order of 25 vol%).

361 In natural systems (i.e. planetesimals in the early solar system), silicate and metal-rich
362 phases fractions have close melting temperatures (~1000°C). Furthermore, the first melt
363 compositions produced upon heating of the metal-rich fraction are sulfur-rich, close to the
364 Fe-FeS eutectic composition. To our best knowledge, a single study reports for
365 measurements of dihedral angles in a three-phase system hosting sulfide melts (Holzheid et
366 al., 2000). As dihedral angle measurement resolution is not specified, these values have to
367 be used with caution. In their study, the $\gamma_{\text{Melt-Ol}}/\gamma_{\text{Melt-Sulfide}}$ ratio is greater than 1, whereas the
368 $\gamma_{\text{Melt-Ol}}/\gamma_{\text{Melt-Ni}}$ is lower than 1 in the case of a pure metal implying that $\gamma_{\text{Melt-Sulfide}} < \gamma_{\text{Melt-Ni}}$. This
369 switch in the value of the ratio $\gamma_{\text{Melt-Fo}}/\gamma_{\text{Melt-Sulfide}}$ compared to $\gamma_{\text{Melt-Fo}}/\gamma_{\text{Melt-Ni}}$ indicates a
370 change in the order of interfacial energies, i.e. of energetic regime, relative to the case of
371 pure metal, which then becomes $\gamma_{\text{Melt-Sulfide}} < \gamma_{\text{Melt-Ol}} < \gamma_{\text{Ol-Sulfide}}$ (Figure 5). The sulfur-rich melt
372 thus appears to have a low interfacial energy with silicate melt and a higher one with olivine
373 so that the contact between sulfur-rich melt and solid silicates tends to be limited. In this

374 case, the sulfide melts produced at low temperature near the eutectic will tend to spread
375 along the surface of solid silicate grains in order to increase the contact surface with the
376 silicate melt, thus having a tendency to interconnect in the presence of a silicate melt
377 (Figure 5).

378 **4.3 Metal-silicate differentiation in planetesimals**

379 Comparing dihedral angle measurements carried out in this study for a pure metal with
380 previous ones for a sulfur-rich phase shows that increasing the sulfur content of the metallic
381 liquid increases its wettability, reducing its dihedral angle and its percolation threshold.
382 Once this threshold exceeded, the sulfur-rich metallic liquid is able to percolate, leaving a
383 metallic residue whose threshold increases due to decreased sulfur content. Therefore, in the
384 absence of a large degree of silicate melting, the complete extraction of metal in a small
385 body remains unlikely.

386 To explore the consequences of a dependence of the interconnection threshold on sulfur
387 content, the evolution of a body heated by decay of ^{26}Al has been modeled. Upon heating,
388 silicate and iron-sulfur begin to melt at similar temperature (see below). The behavior of
389 each subsystem thus has to be modeled as their relative proportions depend on each other.
390 Silicate melt is likely to form an interconnected network even at very low melt fraction (<1
391 vol% estimated from Laporte and Provost, 2000; Wray, 1976), thus it is assumed to be
392 continuously extracted. As the fraction of silicate decreases, that of sulfide melt increases
393 until the interconnection threshold is reached. This behavior depends at the same time on

394 the relation between the sulfur content of the sulfide melt and its interconnection threshold
395 and on the melting curve of silicates.

396 **4.3.1 Modeling the metal-sulfide subsystem**

397 The metallic subsystem is assumed to have an initial composition of H-type ordinary
398 chondrites as they are a possible precursor material of primitive achondrites (Palme et al.,
399 1981). Such a composition corresponds to volumetric phase proportions of 3 vol% troilite
400 and 7 vol% metal (Guignard and Toplis, 2015), or 7 wt% S in the metallic subsystem
401 (Wasson and Kallemeyn, 1988). As temperature rises, removal of silicate melt increases the
402 volume fraction of metal, but the sulfur content of the sulfide melt decreases due to
403 equilibrium melting of the metallic subsystem. The overall effect is an increase of the
404 interconnection threshold of the molten fraction, from a lower limit at the Fe-FeS eutectic (3
405 vol%, Roberts et al., 2007; Yoshino et al., 2003) to an upper limit for pure metal (25 vol%
406 estimated from the large value of nickel dihedral angle measured in the present study, see
407 3.2.3). Between these two endmembers, the sulfur content – interconnection threshold
408 dependence is poorly constrained. For this reason, several different functions have been
409 considered (Figure 6a). Threshold function (a) increases linearly with sulfur content from the
410 Fe-S eutectic composition to pure iron, while other functions investigate non-linear trends.
411 Altogether, they fall into two categories: a first one with with a sharp increase of the
412 threshold upon decreasing sulfur content in the vicinity of the eutectic (threshold functions
413 a and b), and a second one with a smooth increase of the threshold upon decreasing sulfur

414 content in the vicinity of the eutectic (threshold functions c, d and e). The liquidus curve of
415 the Fe-S phase diagram is taken from Buono and Walker (2011) at 1 bar (Figure 7, black
416 curve) and allows plotting the different percolation threshold functions as a function of
417 temperature (Figure 6b, full lines). The percolation thresholds increase with temperature until
418 all the metallic subsystem is entirely molten ($\sim 1400^{\circ}\text{C}$ at 7 wt% of sulfur, dashed black thin
419 vertical line Figure 6b), then the threshold functions reach constant values. In the following,
420 it is considered that once the interconnection threshold reached, the network forming
421 sulfide melt is extracted.

422 Another important issue is the efficiency of draining once the percolation threshold has
423 been reached. Ghanbarzadeh et al. (2017) demonstrated that drainage is affected by an
424 hysteresis effect, where not only the molten fraction exceeding the percolation threshold
425 flows out, but almost all the fluid is lost, except for a small fraction trapped in network
426 irregularities. This amount corresponds to the trapping threshold, which is zero for a
427 dihedral angle less than 60° as in the case of silicate melts, but up to 2 vol% for a dihedral
428 angle of 120° (Ghanbarzadeh et al., 2017). In the present calculation, the trapping threshold
429 is set to 1 vol%.

430 **4.3.2 Modeling the silicate subsystem**

431 The melting curve of the silicate fraction has been calculated using a fractionate melting
432 simulation on the *Rhyolite-Melts* thermodynamic software, with continuous extraction of the
433 melt (Asimow and Ghiorso, 1998; Ghiorso and Gualda, 2015; Ghiorso and Sack, 1995; Gualda

434 et al., 2012). An initial composition of H-type chondrites (Wasson and Kallemeyn, 1988) is
435 also assumed and temperature ranges from 950°C to 2000°C, at 1 bar, along the Iron-
436 Wüstite (IW) buffer. The FeO content of the silicates is calculated from the $\text{FeO}/(\text{FeO}+\text{MgO})$
437 ratio (Wasson and Kallemeyn, 1988, Table 1). The melting curve is displayed in Figure 6c and
438 6d as a black curve. The different kinks in the curve correspond to the complete melting of
439 the different silicate phases. This simulation shows a silicate eutectic around 1000°C.

440 **4.3.3 Metal segregation**

441 Upon removal of the silicate melt produced, the quantity of metal increases (whole
442 metal, i.e. solid iron and sulfide on Figure 6c and sulfide melt on Figure 6d). Once the
443 quantity of sulfide melt reaches its interconnection threshold, all but 1 vol% is assumed to
444 be extracted as explained above. This removal causes the loss of sulfur, changing the
445 composition of the metallic subsystem (Figure 7, color curves for the different threshold
446 functions) if it occurred while there was still some solid iron. In this case, the interconnection
447 threshold is shifted towards higher values; this tendency is represented in Figure 6b (dashed
448 lines) and S2.

449 The different threshold functions described here fall in two categories. In the first case,
450 the interconnection threshold of the sulfide cannot be reached until a large fraction of
451 silicate is melted. For example, threshold function (a) leads to the result that the high
452 interconnection threshold of pure metal is preserved over a wide range of compositions
453 (Figure 6a). In this pessimistic case, the conditions to extract the sulfur-bearing component

454 would require 67% melting of the silicate fraction, corresponding to an extreme temperature
455 of ~1850°C (Figure 6b, 6c and 6d). In the case of threshold function (b) (linear increase with
456 sulfur content), the threshold is constantly higher than the volume fraction of sulfide melt
457 over a wide range of temperatures (Figure 6b, 6c and 6d), such that the threshold is not
458 reached before extraction of 55% of the silicates, i.e. a temperature of 1600°C. The same
459 sort of behavior is also true for threshold function (c), that is a non-linear function of S-
460 content, with lower interconnection threshold at high S-content. In this case, the onset of
461 drainage occurs at 13 vol% of molten sulfide and a degree of silicate melting of 30 wt%, i.e.
462 1450°C (Figure 6b, 6c and 6d)., For the three cases described above (threshold functions (a),
463 (b) and (c)), sulfide melt extraction requires the removal of large volume of silicate melt and
464 temperatures above 1420°C, meaning that the metallic subsystem is fully molten and its
465 extraction takes place in a single event, leaving only a small residual trapped fraction of 1
466 vol%. Such a scenario appears unlikely given that the loss of silicate melt also takes away
467 the principal heat source, ^{26}Al that is transferred to the melt by plagioclase melting near
468 1200°C. In the case of fractional melting assumed here, the ^{26}Al heat sources will thus be
469 totally extracted for a melting degree no higher than 15%. However, models on the
470 percolation process in Vesta (Mizzon, 2015) show that silicate melt migration is not
471 instantaneous. Even though the heat source is transferred towards the surface of the body,
472 the degree of mantle melting can be as high as 50% due to time-scale of melt extraction.

473 For the second family of cases, threshold functions (d) and (e) allow interconnection and
474 extraction of the sulfide melt at lower temperatures, i.e. at lower silicate melting degrees
475 and below the liquidus of the metallic subsystem. In details, function (d) is at first glance
476 similar to (c), but it yields a very different extraction behavior (Figure 6b, 6c and 6d). In this
477 case, a first episode of drainage occurs at 3 vol% of liquid sulfide, after extraction of 19% of
478 silicate melt (1250°C). This concerns 2% of the total volume, corresponding to 20% of the
479 metallic phase. This loss of iron-sulfide liquid reduces the sulfur content of the residual
480 metallic subsystem by two-thirds of its initial value, thus raising the interconnection
481 threshold to 18% (Figure 6b and 7). Both factors inhibit a possible second extraction of
482 sulfide melt that would occur at a silicate melting degree of 65% and a temperature of
483 1800°C (Figure 6b, 6c and 6d), conditions that cannot be reached easily for the
484 aforementioned reasons. As a consequence, only the first episode of loss of molten sulfide
485 remains plausible. In this case, it will leave behind a material with a metal volume fraction
486 close to the initial state, i.e. 10 vol%, but depleted in sulfur (~2.5 wt%, Figure 7), and a
487 silicate highly depleted in its feldspar and clinopyroxene components. Function (e) assumes
488 that the interconnection threshold of the eutectic is preserved for a wide range of sulfur
489 contents and allows three successive extractions at 1220°C, 1380°C and 1600°C
490 corresponding respectively to 17%, 24% and 50% of silicate partial melting (Figure 6b, 6c
491 and 6d). These stages result in a metal volume fraction of 10%, 8% and 1% (trapped
492 fraction) respectively, the final episode of drainage taking place after complete fusion of the

493 metal. During these stages the sulfur content decreases to 2.3 wt% and then to 1 wt%
494 (Figure 7). Interestingly, function (e) belongs to both threshold function categories, since it
495 allows partial metal extraction at moderate silicate partial melting (17%), and then complete
496 drainage at a higher but perhaps still possible degree of silicate melting (50%).

497 **4.3.4 Comparison with experimental data and primitive achondrites**

498 The present model indicates that the S-content dependence of the interconnection
499 threshold plays a critical role on the drainage of sulfur-bearing liquids. In the absence of
500 direct experimental determination concerning the effect of S-content, we turn to
501 experiments in similar systems and data from natural samples. For example, experiments
502 conducted by Rose and Brenan (2001) addressed the effect of oxygen on the wettability of
503 metallic melts. They observed a sharp decrease in the dihedral angle from 0 to 5 mol% of
504 oxygen, followed by a smooth decreasing linear trend at higher oxygen content, a behavior
505 similar to that described by functions (c), (d) or (e). Holzheid et al. (2000) built a data
506 compilation addressing the effect of sulfur, but it lacks data on pure metal. Adding the data
507 from the present study, a sharp decrease of the interconnection threshold is observed at
508 low sulfur content, followed by a smooth decreasing linear behavior, again similar to curves
509 (c), (d) and (e). In light of these considerations, it is concluded that this sort of dependence
510 is the most plausible.

511 In turn, this sheds new light on the processes experienced by primitive achondrites and
512 may potentially explain their composition. These meteorites are widely accepted to

513 represent partial melting residues for both metallic and silicate subsystems. Sulfur contents
514 of the Fe-FeS system is plotted in Figure 7 for different primitive achondrites. Note that the
515 composition can be highly heterogeneous in the same meteorite, from one section to
516 another: for instance Acapulco displays S-contents ranging from ~6 wt% up to ~15 wt%
517 (Guignard and Toplis, 2015; Palme et al., 1981; Zipfel et al., 1995). Primitive achondrites can
518 nonetheless be assigned to two categories following their S-content: they are either
519 depleted or enriched relative to the H-chondrite primitive material (Figure 7). Depletion can
520 be due to the extraction of a sulfide melt, whose composition is different from that of the
521 bulk metallic subsystem. In a similar way, S-enriched meteorites can be viewed as regions
522 that have locally concentrated sulfide melts draining down from upper parts of the parent
523 body. Indeed, it should be appreciated that differentiation at moderate melt fraction
524 operates through liquid migration inside a porous solid matrix. This migration is affected by
525 compaction waves that form alternating horizons enriched and depleted in liquid (Šrámek et
526 al., 2012) that may freeze if the heat source (i.e. ^{26}Al content) is insufficient to allow
527 complete separation. S-enriched and S-depleted meteorites can thus sample different
528 horizons of the same planetesimal. This feature may be simply understood in the case of a
529 S-dependent threshold corresponding to function (d) or (e). Recent studies on Hf-W
530 systematics (Kruijjer et al., 2014) also support the idea that different sulfide melts extraction
531 events may occur during differentiation. More precisely, data on IVB iron meteorites suggest
532 two segregation steps: a first one with an early extraction of a S-rich liquid, and a second

533 one, later, with an iron-rich melt. However, the present study outlines that the extraction of
534 a silicate melt is a necessary condition to allow the metallic melt fraction to exceed its
535 interconnection threshold.

536 Finally, the formation of a small core with a heterogeneous sulfur content is a natural
537 outcome of the present model depending on the temperature of extraction, as melts with
538 various compositions are extracted. This model thus provides a plausible explanation for the
539 diversity of sulfur content in iron meteorites, even though one should keep in mind that
540 these meteorites most likely formed by larger degrees of melting and are expected to have
541 lost some sulfide due to the sulfur volatility and the mechanical weakness of troilite.

542 **5 Conclusion**

543 Laboratory experiments were performed to study the local geometry of a three-phase
544 system composed of forsterite, silicate melt and nickel grains relevant to partially
545 differentiated meteorites. The dependence of equilibrium geometries (dihedral angles) on
546 interfacial energies was also considered from a theoretical point of view. Finally, the
547 evolution of the metal fraction in a planetesimal was modeled assuming an interconnection
548 threshold that is a variable function of S-content. The main conclusions can be summarized
549 as follows:

- 550 • From an experimental and analytical point of view, a resolution of 30 nm per
551 pixel is necessary for accurate measurements of dihedral angles, in particular the

552 complex shape of forsterite grains that precludes simple prediction of the
553 different interconnection thresholds of the system. A theoretical approach shows
554 that different energetic regimes exist and that the accurate measurement of
555 dihedral angles is needed to predict the geometry.

556 • Fe-FeS eutectic melt (first metallic liquid to form) and pure metal (last metallic
557 liquid to form) belong to two distinct interfacial energy regimes. The comparison
558 between the equilibrium geometries in a system containing Fe-FeS eutectic melt
559 shows that the sulfide – silicate melt interfacial energy is energetically favorable
560 when the metallic melt is sulfur-rich, meaning that such liquids will spread along
561 the faces of silicate crystals, decreasing the interconnection threshold of the
562 metallic melt. On the other hand, in the case of pure metal, grains remain largely
563 spherical, increasing the interconnection threshold.

564 • The variation of sulfur content of primitive achondrites from one sample to the
565 other is best explained when the interconnection threshold significantly decreases
566 at low sulfur content and remains close to the value of the Fe-FeS eutectic for a
567 wide range of compositions at higher S contents. Different sulfide melt
568 extractions are then possible at temperatures of 1200-1400°C, requiring ~25 vol%
569 of silicate melt. These conditions can be reached even if the mobility of the heat
570 source is taken into account. Successive extraction events provide a good
571 explanation for the variability of sulfur content observed in primitive achondrites,

572 which are either representative of a region that experienced sulfide extraction or
573 from a region that accumulated sulfide melt from the outer parts of the body.

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582

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735

736 **Figure 1:** SEM images representative of the different samples. White: nickel; dark grey:
737 forsterite and light grey: silicate melt. (a.) 47.5:47.5:5 sample at x1000 (94 nm per pixel),
738 (b.) 32.5:62.5:5 sample at x1000 (94 nm per pixel), (c.) 20:75:5 at x1000 (94 nm per pixel),
739 (d.) 75:20:5 sample at x1000 (94 nm per pixel), (e.) 20:75:5 sample at x3000 (30 nm per
740 pixel), (f.) 75:20:5 sample at x3000 (30 nm per pixel), (g.) 20:75:5 sample at x10 000 (9.4
741 nm per pixel), (h.) 75:20:5 sample at x10 000 (9.4 nm per pixel). Images on (g.) and (h.)
742 highlight the hook shape of the silicate melt toward the apex. This kind of structure is
743 typically of 100 nm at half-height of the hook, requiring a resolution threshold of 30 nm
744 per pixel to have a proper imaging.

745
746 **Figure 2:** Measured median dihedral angle as a function of pixel size (in nm) for (a)
747 silicate melt, (b) forsterite, (c) nickel and merged dihedral angles distributions at different
748 pixel sizes: (d) 376 nm per pixel, (e) 94 nm per pixel and (f) 30 and 9.4 nm per pixel.
749 Circle color symbols on (a), (b), (c) correspond to different samples, black circle
750 represents the median value of all datasets merged at a given magnification and gray
751 lines are guides for the eyes. These three diagrams show that the dihedral angle reaches
752 steady value at a pixel size of 30 nm per pixel, corresponding to the resolution at which
753 the fine microstructure is entirely imaged. Scatter between different samples at lower
754 resolution is due to measurement artifacts, as this small-scale geometry is not entirely
755 imaged. Step curves on (d), (e) and (f) correspond to the dihedral angle distribution,
756 curly curves correspond to Gaussian fits, single Gaussian function for (d) and (e) and
757 triple Gaussian function for (f); black dotted lines represent the median value of the
758 distributions. The agreement between data and triple Gaussian fit is shown in (g) for the
759 silicate melt distribution and in (h) for the olivine distribution. Orange, green and blue
760 dotted vertical lines correspond to median values of each subset of dihedral angle.
761 Increasing the resolution makes the distribution more asymmetric, meaning that different
762 populations of dihedral angle exist, representative of the forsterite crystalline anisotropy
763 at microscale.

764
765 **Figure 3:** Schematic view of a triple junction between the silicate melt, a forsterite grain
766 and a nickel grain. Dihedral angles θ_1 , θ_2 and θ_3 are related to interfacial energies γ_{12} , γ_{13}
767 and γ_{23} when textural equilibrium is achieved, following equation 2.

768

769 **Figure 4:** (a) Ternary diagram showing the inner triangle of possible space given the
770 conditions of $\theta > 0^\circ$ and $\theta < 180^\circ$. (b) Reduced ternary diagram showing the different
771 energetic regimes. Full lines separate these regimes and represent equality between two
772 interfacial energies. Sketches explaining the geometry in remarkable points of the ternary
773 diagram are represented in (c) and (d). Two end-member cases are considered: (c) metal
774 is the minor phase, (d) silicate melt is the minor phase. If one angle is equal to 180° , the
775 phase either forms a sphere or a film. When the dihedral angle is equal to 0° , the phase
776 is highly wetting, its contact is punctual or inexistent, and this phase prevents the two
777 others from being in contact with each other.

778
779 **Figure 5:** Ternary diagram of the dihedral angles with the different energetic regimes.
780 Purple symbols represent data from this study. Pentagon symbols stand for the different
781 populations of dihedral angles taking into account forsterite crystalline anisotropy
782 (merged data at resolutions of 30 and 9.4 nm per pixel). Grey symbols correspond to the
783 data from (Holzheid et al., 2000). Error bars are smaller than the symbols. Sketches show
784 the geometry of the three phases according to their dihedral angles. Color code is the
785 same as in Figure 3, except for sulfide, in red. Data with pure metal fall in the $\gamma_{12} < \gamma_{13} <$
786 γ_{23} ($\gamma_{\text{Melt-OI}} < \gamma_{\text{Melt-Ni}} < \gamma_{\text{OI-Ni}}$) domain, even when crystalline anisotropy is taken into account
787 (shaded pentagons), which is clearly different from a system with sulfide: $\gamma_{13} < \gamma_{12} < \gamma_{23}$
788 ($\gamma_{\text{Melt-Sulfide}} < \gamma_{\text{Melt-OI}} < \gamma_{\text{OI-Sulfide}}$).

789
790 **Figure 6:** Evolution of the metal volume fraction of a silicate-metal mixture as a function
791 of temperature. Silicate melt is assumed to be continuously extracted so that the metal
792 fraction increases with the melting degree of silicate. The metallic melt is completely
793 drained when its percolation threshold is reached, except a trapped fraction set to 1
794 vol%. The percolation threshold is a function of the sulfur content. (a) Different
795 interconnection threshold evolutions as a function of sulfur content of the sulfide melt.
796 (b) Percolation threshold functions as a function of temperature. Full lines are for
797 simulations that experienced sulfide melt extraction once all the iron is molten, while
798 dashed lines represent the case where the extraction occurred when a solid iron residue
799 was present. (c) Evolution of the metal volume fraction (solid iron and sulfide melt) as
800 the silicate is extracted through fractionated melting. (d) Evolution of the sulfide melt
801 volume fraction as a function of temperature. In (c) and (d) the black line represents the
802 volume of silicate melt extracted as a function of temperature, the shaded area
803 indicating the residual silicate. The different kinks in the silicate melting curves represent
804 the disappearance of mineral species, arrows pointing them out. Color lines draw the
805 volume fraction of the metallic subsystem in (c) and of the sulfide melt in (d) for the
806 different percolation threshold functions depicted in (a) and (b).

807

808 **Figure 7:** Evolution of the sulfur content of metal plotted on the phase diagram of the
809 Fe-FeS system. Color dots represent the composition of the extracted melt, following the
810 color code for the different threshold functions of Figure 6a. Color bars represent the
811 composition of different primitive achondrites, sulfur enriched ones in pink (Acapulco
812 (Guignard and Toplis, 2015; Zipfel et al., 1995) and TIL99002 (Patzner et al., 2004)) and S-
813 depleted ones in yellow (Acapulco (Palme et al., 1981), Lodran (Bild and Wasson, 1976;
814 Guignard and Toplis, 2015) and NWA 725 (Patzner et al., 2004)). The discrepancy in the
815 sulfur content of the different primitive achondrites can be accounted for given that the
816 evolution of the interconnection threshold is close to threshold functions (d) or (e),
817 allowing the fractionation of sulfur within the planetesimal.

818

819

820 **Table 1: Composition in wt% of the starting materials and composition of the different phases after**
821 **experiments. Error bars, expressed as 2σ standard deviation, were estimated based on repeated**
822 **measurements of at least 30 points for each phase.**

Phase composition (wt%)	MgO	SiO ₂	CaO	Al ₂ O ₃	NiO	Sum
Commercial Fo	57.55 ± 0.78	43.03 ± 0.55	0.55 ± 0.16	0.00	0.00	101.17
Silicate melt aimed composition	22.92	45.49	14.55	17.04	0.00	100.00
Starting silicate melt	23.83 ± 0.32	44.72 ± 0.84	14.70 ± 0.31	16.75 ± 0.49	0.00	100.00
Fo in Fo:Melt:Ni (20:75:5)	54.68 ± 0.58	42.50 ± 0.52	0.50 ± 0.05	0.20 ± 0.07	2.12 ± 0.73	100.00
Fo in Fo:Melt:Ni (47.5:47.5:5)	54.78 ± 0.45	42.47 ± 0.53	0.50 ± 0.06	0.20 ± 0.08	2.04 ± 0.054	100.00
Silicate melt in Fo:Melt:Ni (20:75:5)	21.32 ± 0.57	44.58 ± 0.22	15.23 ± 0.44	18.26 ± 0.33	0.60 ± 0.26	100.00
Silicate melt in Fo:Melt:Ni (47.5:47.5:5)	21.43 ± 0.29	43.94 ± 0.19	15.50 ± 0.13	18.53 ± 0.15	0.60 ± 0.11	100.00

823
824 **Table 2: Median dihedral angles (in °) at the triple junctions for different magnifications and all samples. The**
825 **merged sections correspond to all dataset put together. In each column the value of the dihedral angle is**
826 **reported with its 2SE ($2SE = \frac{2SD}{\sqrt{N-1}}$ where SD is the standard deviation and N the number of measurements)**
827 **uncertainty.**

Composition		×250	×1,000	×3,000	×10,000
20:75:5	Fo	126.7 ±11.0	149.6 ±5.1	161.1 ±5.0	164.7 ±6.3
	melt	62.6 ±1.1	37.4 ±5.5	24.1 ±6.1	21.2 ±5.2
	Ni	170.9 ±0.6	175.3 ±2.3	177.3 ±4.0	179.2 ±4.2
47.5:47.5:5	Fo	126.2 ±1.5	145.9 ±6.1	166.8 ±7.2	164.4 ±4.3
	melt	63.4 ±1.7	38.4 ±6.3	19.3 ±6.8	18.3 ±7.4
	Ni	167.7 ±0.8	174.2 ±2.6	176.6 ±3.0	180.0 ±5.1
62.5:32.5:5	Fo	126.9 ±1.5	150.2 ±5.1	156.2 ±5.9	157.6 ±6.2
	melt	80.0 ±1.6	46.9 ±5.2	27.8 ±5.6	24.5 ±5.5
	Ni	155.6 ±0.8	165.8 ±1.8	175.0 ±1.7	175.6 ±2.3
75:20:5	Fo	122.4 ±1.1	145.5 ±4.5	162.6 ±5.5	163.6 ±9.4
	melt	84.3 ±1.2	50.8 ±4.6	26.5 ±5.0	22.6 ±6.6
	Ni	155.7 ±0.5	164.5 ±1.5	174.1 ±2.2	176.2 ±4.8
Merged	Fo	125.6 ±1.2	148.4 ±2.6	161.1 ±3.0	161.7 ±3.8
	melt	70.5 ±1.4	42.2 ±2.7	24.7 ±2.9	21.4 ±3.1
	Ni	162.3 ±0.7	168.8 ±1.1	175.3 ±1.3	176.9 ±1.8

828
829 **Table 3: Dihedral angles of the different phases when crystalline anisotropy is considered and corresponding**
830 **interfacial energies. * correspond to values calculated from dihedral angle data. Uncertainties are 2SE**
831 **calculated using the derivative method.**

Populations	Phase	Dihedral angle (degree)	Interfacial energy ratios
x250	Fo	125.6 ±1.2	$\gamma_{\text{Melt-Fo}}/\gamma_{\text{Melt-Ni}}$ 0.374 ±0.016
	Melt	70.5 ±1.4	$\gamma_{\text{Melt-Fo}}/\gamma_{\text{Fo-Ni}}$ 0.323 ±0.013
	Ni	162.3 ±0.7	$\gamma_{\text{Melt-Ni}}/\gamma_{\text{Fo-Ni}}$ 0.862 ±0.015
x1000	Fo	148.4 ±2.6	$\gamma_{\text{Melt-Fo}}/\gamma_{\text{Melt-Ni}}$ 0.372 ±0.044
	Melt	42.2 ±2.7	$\gamma_{\text{Melt-Fo}}/\gamma_{\text{Fo-Ni}}$ 0.290 ±0.031
	Ni	168.8 ±1.1	$\gamma_{\text{Melt-Ni}}/\gamma_{\text{Fo-Ni}}$ 0.781 ±0.070
Subset 1	Fo	173.1 ±0.7	$\gamma_{\text{Melt-Fo}}/\gamma_{\text{Melt-Ni}}$ 0.358 ±0.104
	Melt	15.2 ±1.1	$\gamma_{\text{Melt-Fo}}/\gamma_{\text{Fo-Ni}}$ 0.265 ±0.077
	Ni	176.1 ±1.1	$\gamma_{\text{Melt-Ni}}/\gamma_{\text{Fo-Ni}}$ 0.741 ±0.071

Subset 2	Fo	162.2	±1.5	$\gamma_{\text{Melt-Fo}}/\gamma_{\text{Melt-Ni}}$	0.140	±0.040
	Melt	35.5	±1.2	$\gamma_{\text{Melt-Fo}}/\gamma_{\text{Fo-Ni}}$	0.125	±0.035
	Ni	176.1	±1.1	$\gamma_{\text{Melt-Ni}}/\gamma_{\text{Fo-Ni}}$	0.893	±0.050
Subset 3	Fo	137.5	±2.7	$\gamma_{\text{Melt-Fo}}/\gamma_{\text{Melt-Ni}}$	0.093	±0.026
	Melt	52.9	±3.2	$\gamma_{\text{Melt-Fo}}/\gamma_{\text{Fo-Ni}}$	0.087	±0.025
	Ni	176.1	±1.1	$\gamma_{\text{Melt-Ni}}/\gamma_{\text{Fo-Ni}}$	0.943	±0.060
Merged	Fo	161.3	±2.4	$\gamma_{\text{Melt-Fo}}/\gamma_{\text{Melt-Ni}}$	0.214	±0.066
	Melt	23.3	±2.1	$\gamma_{\text{Melt-Fo}}/\gamma_{\text{Fo-Ni}}$	0.174	±0.052
	Ni	176.1	±1.1	$\gamma_{\text{Melt-Ni}}/\gamma_{\text{Fo-Ni}}$	0.811	±0.121
Literature data representative of natural samples				Interfacial energy ratios		
(Ballhaus and Ellis, 1996)	Sulfide	68 to 86		$\gamma_{\text{O}^{\text{I}}-\text{FeS}^*}$	1.463 to 1.658	
	Two-phase system	FeNi alloy	113 to 115	$\gamma_{\text{O}^{\text{I}}-\text{FeNi}^*}$	1.075 to 1.104	
(Holzheid et al., 2000)	OI	136	±5	$\gamma_{\text{Melt-Fo}}/\gamma_{\text{Melt-FeS}^*}$	1.221	±0.129
	melt	102	±5	$\gamma_{\text{Melt-Fo}}/\gamma_{\text{Fo-FeS}^*}$	0.867	±0.050
	Three-phase system	Sulfide	122	±5	$\gamma_{\text{Melt-FeS}}/\gamma_{\text{Fo-FeS}^*}$	0.710

832













