1 # The Fate of Carbon during Earth's Core–Mantle Differentiation

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11

12 **Abstract**

13 **Carbon is an essential element for the existence and evolution of life on Earth, constitutes**

14 **up to 50% of dry biomass, and is likely a requirement for all life in the universe. Its high**

15 **abundance in Earth's crust and mantle (the Bulk Silicate Earth, BSE) is surprising**

16 **because carbon is strongly siderophile (metal-loving) and should have segregated almost**

17 **completely into Earth's core during accretion[1–3]. Estimates of the concentration of carbon**

18 **in the mantle lie mostly in the range of 80–120 ppm[4,5], which is much higher than expected**

19 **based on simple models of core–mantle differentiation[2,6,7]. Here we show through**

20 **experiments at 49–71 GPa and 3600–4000 K that carbon is significantly less siderophile**

21 **at such conditions than at the low pressures (≤ 3 GPa) of previous studies[1–3,8]. We derive**

22 **a new parameterization of the pressure–temperature dependence of the metal–silicate**

23 **partitioning of carbon and apply this in a state-of-the-art model of planet formation and**

24 **differentiation[9,10] that is based on astrophysical N-body accretion simulations. Results**

25 **show that BSE carbon concentrations increase strongly starting at a very early stage of**

Earth's accretion and, depending on the concentration of carbon in accreting bodies, can easily reach or exceed estimated BSE values. In contrast, simple models of "continuous core formation"[11–13] require all BSE carbon to be accreted after core formation ended, but this is not consistent with astrophysical models of accretion.

The extents to which the volatile contents of terrestrial planets were determined by the processes of accretion and core formation, impact-driven volatile loss, and addition of a late veneer subsequent to core formation are highly controversial[9,14–17]. The Earth is considered to have formed from bodies of chondritic composition, although it is unlikely that any particular chondrite group constituted the sole building blocks of the planet[18]. Different classes of chondrites have varying carbon concentrations, but importantly, such concentrations are all high, at the weight percent level[19]. This is far in excess of the concentration of carbon present in the Bulk Silicate Earth (BSE) today, which is currently estimated to be 80–120 ppm[4,5] or possibly as high as 765 ± 300 ppm[20]. Additionally, the metal–silicate partition coefficient of carbon at low pressures and temperatures (≤ 13 GPa and ≤ 2000 K) is very high ($D > 1000$; see D defined in Eq. 2 and ref.[1–3,21]). Consequently, almost all carbon should have partitioned into the core during its formation, leaving the mantle almost completely depleted in this element. Several explanations have been proposed for the BSE C concentration, including crystallization of C-rich phases (e.g. diamond) from a C-saturated magma ocean, imperfect metal–silicate equilibration during core formation, accretion of a "late veneer" after the end of core formation, and carbon outgassing from the core[1,3,22].

Here we present the results of experiments that determine the metal–silicate partition coefficient of carbon at conditions that are likely similar to that of metal–silicate equilibration/segregation during core formation[23], and that are at much higher pressures and temperatures than those of previous studies. We use the results in a state-of-the art model that combines accretion and

51   core–mantle differentiation[9,10], and so we show that the present BSE carbon concentration is

52   the direct consequence of core formation.

53   We performed laser-heated diamond anvil cell (LH-DAC) experiments to reproduce the

54   pressures and temperatures of the putative conditions of Earth's core-mantle differentiation in

55   the laboratory. We performed experiments at 49–71 GPa and 3600–4000 K for a few tens of

56   seconds using a double-sided laser-heated system (see Methods section for details). After

57   melting and quenching at high pressure, our recovered samples consisted of a central metallic

58   sphere surrounded by quenched silicate glass, as observed in previous similar studies[24–26] (Fig.

59   1a). Samples were prepared for analysis using a Focused Ion Beam system and analysed for

60   major elements using Electron Probe Microanalysis (EPMA see Methods section). To measure

61   the concentrations of carbon in the quenched silicate melt, we used NanoSIMS, for which we

62   synthesized appropriate standards (the carbon contents of which were analysed by FTIR; see

63   the Methods section).

64   We calculated the oxygen fugacities of our experiments relative to the iron-wüstite redox buffer

65   (ΔIW), based on the assumption of ideal mixing of both phases (as justified by the high

66   temperatures):

67     $\Delta\text{IW} = 2\log\dfrac{X_{FeO}}{X_{Fe}}.$ (1)

68   Oxygen fugacities of our experiments lie between -0.9 and -1.5 log units relative to the IW

69   buffer (Table 1). The concentration of carbon in the metal lies in the range from 5 to 9.4 wt.%

70   and in the quenched silicate melt varies from 545 to 2800 ppm. The concentrations of carbon

71   in the quenched silicate melt are much higher than concentrations observed in low P–T

72   experiments which are typically 10–200 ppm[1–3,8]. Thus, metal–silicate partition coefficients for

73   carbon are 24−166 (Table 1) and are 1–2 orders of magnitude lower than those determined in

74   previous low-pressure studies.

75   In order to understand the metallic phase better, we obtained images with SEM and TEM of the

76   sample BAS C 39 (Fig. 1), and we performed electron diffraction analyses that show the

77   presence of stoichiometric Fe_7C_3 inclusions in the quenched metal (Fig. 1b). To further

78   investigate the quench textures (Fig. 1c), we performed scanning TEM-energy dispersive

79   spectroscopy (EDS) mapping to identify Si-O-rich exsolved inclusions in the metal phase (Fig.

80   1d) along with a C-rich domain that could be an exsolved diamond, as also observed in previous

81   similar experiments[27].

82   To understand the effects of pressure and temperature on the metal–silicate partitioning of

83   carbon, we assembled a dataset for our model from previous studies to supplement our new

84   results (Fig. 2). However, only some published experiments are relevant to core formation

85   because prior studies were often focussed on the effects of silicate melt composition[2], fO_2 [28,29],

86   and the interaction of carbon with elements in the metal[7]. The partitioning of carbon is affected

87   by the ratio of non-bridging oxygens to tetrahedral cations[2] (NBO/T), so we only included data

88   from the literature for which NBO/T lies in a narrow range (0.5–1.5); the range for our samples

89   is 0.56–1.34. We also only included data from previous low P–T studies with low

90   concentrations (< 1 wt.%) of light elements (S, N, O, Si) in the metal because the interaction

91   between carbon and other light elements may be significant[6] but is poorly constrained. As a

92   consequence, we also excluded very low fO_2 data (ΔIW < -3), as they are often associated with

93   high concentrations of Si in the metal. On the other hand, our high P-T samples contain

94   significant concentrations of oxygen and silicon in the metallic phase, as is typical for high

95   temperature experiments[24,25]. By combining these two datasets, we thereby empirically include

96   the interaction effects of these light elements in the regression presented below, because the

97   concentrations of O and Si in metal are generally low during core formation at low P–T

98   conditions[11,31] consistent with the data[1-3,8] selected from previous studies, whereas at high P–T

99   conditions such concentrations become high[30,31], which is consistent with the present study.

100   Interactions of carbon with oxygen and silicon in the metal of our experiments might be at least

101   partly responsible for the scatter of our data (Fig. 2). Finally, we did not use literature data for

102   which the silicate phase did not quench to a glass, because the quality of SIMS analyses may

103   be compromised[1]. In Fig. 2, we present a comparison of our data together with the selected data

104   from the literature as a function of temperature and pressure.

105   The metal–silicate partition coefficient of carbon (D_C) is calculated as:

106    $$D_C = \frac{X_C^{metal}}{X_{(CO3)2-}^{silicate}},\qquad(2)$$

107   where X is the mole fraction of the element in the phase of interest. D is a function of several

108   parameters, including pressure (P), temperature (T), and oxygen fugacity.

109   To remove the effect of oxygen fugacity on D values, we use the distribution coefficient K_D:

110    $$K_D = \frac{D_C}{(D_{Fe})^{n/2}},\qquad(3)$$

111   where n is the valence of C when dissolved in silicate liquid, which is 4 for carbon in this case[32].

112   Based on the aforementioned data selection, we fitted previously– published data[1–3,8] along with

113   our new results to derive the following expression:

114

115    $$\log K_D = -1(\pm0.5) + \frac{4842(\pm920)}{T} + 31(\pm19)\frac{P}{T}.\qquad(4)$$

116

117   This relation shows that carbon becomes less siderophile with increasing depth along a magma

118   ocean geotherm, and predicts K_D values fairly well for both low and high P–T experiments (see

119   Extended Data Fig. S3).

120   We used the parameterization of Eq. 4 in a combined model of accretion and core–mantle

121   differentiation[10]. This model is based on N-body accretion simulations that typically start with

122   80–220 Mars-size embryos (initially located between 0.7 and 3.0 AU) that are embedded in a

123   protoplanetary disk consisting of a few thousand much smaller planetesimals that are initially

124 distributed over a heliocentric distance of 0.7 to ~ 9.5 AU. In the accretion simulations, planets

125 typically grow from the starting embryos through accretional collisions with other embryos and

126 planetesimals. Each collision potentially involves a core formation event during which accreted

127 metal equilibrates with silicate liquid in a magma ocean and then segregates to the proto-core.

128 By modelling metal–silicate equilibration using a mass balance approach, the evolving

129 compositions of the mantles and cores of all accreting bodies are tracked. The approach requires

130 that the compositions of all starting bodies are defined, with refractory element compositions

131 matching CI chondrites. An oxidation gradient in the protoplanetary disk defines the oxygen

132 content of starting bodies, which is the main compositional variable. Embryos and

133 planetesimals that formed within ~1 AU of the Sun are highly reduced but with increasing

134 heliocentric distance are increasingly oxidized[9,33]. Planetesimals originating from beyond ~ 4.5

135 AU are fully oxidized, contain no metal, and have an H_2O content of ~ 20 wt.%. An important

136 feature of the model is that accreted metal only equilibrates with a small fraction of the silicate

137 mantle – which is quantified using a hydrodynamic model of the interaction of metal and silicate

138 as material from the impactor's core sinks in a magma ocean[34].

139 Here we use the "Grand Tack"[35,36] accretion simulation "4:1-0.5-8"[9] with the same model

140 parameters as in our previous study[10]. We first assume that carbon is accreted to Earth only in

141 bodies that originate beyond 4.5 AU and that their average C concentration is 1.6 wt.% (an

142 approximately mean carbonaceous chondrite (CI, CM, CO and CV) value). The mantle C

143 abundance increases throughout accretion from a very early stage and finally reaches the BSE

144 value of 80–120 ppm (Fig. 3). However, in this model the final concentration of C in the core

145 is 47 ppm which means that the carbon concentration of the bulk Earth is only 83 ppm, i.e. far

146 below the lowest estimate of 520 ppm[20]. We have therefore developed a scenario that results in

147 a bulk concentration of ~520 ppm. We assume that bodies from beyond 4.5 AU contain on

148 average 1.6 wt.% carbon and that C concentrations in bodies originating at < 4.5 AU decrease

149   along a linear gradient to reach a value of zero at a heliocentric distance of 1.88 AU. This is

150   based on the condensation of volatile elements such C increasing with heliocentric distance as

151   temperatures decrease and is analogous to a concentration gradient derived for sulfur[10]. The

152   model then predicts a final mantle C concentration of 314 ppm (Fig. 3), a core concentration of

153   950 ppm and a bulk Earth concentration of 518 ppm. This calculation is not unique because the

154   concentration (1.6 wt.% C) and distance (1.88 AU) parameters are of course correlated.

155   Furthermore, if water-rich outer solar system planetesimals have concentrations of C exceeding

156   1.6 wt.%, then it is possible to obtain mantle concentrations consistent with the estimate of 765

157    $\pm$ 300 ppm[20] because C delivered to Earth's mantle is inefficiently extracted by subsequent core

158   formation similar to S and the HSEs[10], as explained below. In summary, the final mantle

159   abundance of Earth exceeds 200 ppm for all plausible C concentrations of the initial bodies in

160   the protoplanetary disk.

161   A BSE carbon concentration of > 200 ppm is considerably higher than the estimate of 100 ± 20

162   ppm[4,5]. If the 100 ± 20 ppm value is correct, an additional process, that is not included in our

163   model, may have transported excess carbon to the core. There is experimental evidence that the

164   pressure-induced disproportionation of $Fe^{2+} \rightarrow Fe^{3+} + Fe$ in deep magma oceans causes the

165   formation of dispersed Fe metal droplets[37,38]. The gravitational segregation of these metal

166   droplets could have transported the excess carbon to the core, leaving ~100 ppm in the mantle

167   and resulting in 0.14 wt.% C in the core.

168   There are two critical features of the accretion/differentiation model that explain why the BSE

169   carbon concentration is not efficiently extracted during core formation in spite of its siderophile

170   behaviour. First, bodies originating from the outer solar system are water ice-bearing and so

171   fully oxidize and contain no metal. Thus, there is no core formation event when they are

172   accreted and the delivered C remains in the magma ocean/mantle. Second, when differentiated

173   metal-bearing bodies are accreted, the metal of the impactor only equilibrates with a fraction of

174 the target's mantle, of which the value depends on the size of the impactor's core and the depth

175 of the magma ocean[9,34]. The fractions of the mantle that equilibrate with metal in the present

176 simulation are 0.16 to 2.5% for planetesimal impacts and 2.6 to 9.9% for embryo impacts.

177 Consequently, core formation events are extremely inefficient at removing carbon from the

178 bulk of the mantle and transferring it to the core. These two critical features are absent in the

179 models of "continuous core formation" that are currently applied in many studies of Earth's

180 differentiation[12,13]; the result of a continuous core formation model for carbon is presented in

181 the supplementary information for comparison. In such models all accreted material is assumed

182 to contain metal and it is assumed (unrealistically) that all accreted metal equilibrates

183 chemically with the entire Earth's mantle. Consequently, such models predict that essentially

184 the entire BSE carbon budget has to be delivered during late accretion, i.e. after core formation

185 has ended (see supplementary Information and Extended Data Fig. S3). We know of no

186 astrophysical accretion simulations that would support this scenario which requires the

187 accretion of carbon-poor material during core formation and carbon-rich material afterwards

188 during late accretion.

189

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290

291 **Table**

292 *Table 1: Experimental conditions of carbon partitioning experiments.*

Run #	BAS C 50	BAS C 60	BAS C 39	BAS C 42
P (GPa)	60	71	49	56
T (K)	3800	4000	3600	3700
ΔIW	-1.06	-1.2	-0.89	-1.51
D_C	149$\pm$7	69$\pm$14	24$\pm$1	166$\pm$6
K_D	12.9$\pm$1	5 $\pm$ 1.4	3.0 $\pm$ 1.0	2.9 $\pm$ 1.1
nbo/t	0.95	0.58	1.34	0.56

293 ΔIW is the calculated fO_2 relative to the IW buffer (see text for details).

294 D_c stands for the metal–silicate partitioning of carbon (mol.% of carbon in the metal / mol.%

295 carbon in the silicate).

296 K_D is the distribution coefficient where $K_D = \frac{D_C}{(D_{Fe})^{n/2}}$, nbo/t is the number of non-bridging

297 oxygen atoms over tetrahedrally coordinated cations.

298 The uncertainties on temperature and pressure measurements are estimated to be about 300 K

299 and 5 GPa respectively.

300

301   **Figures**

302
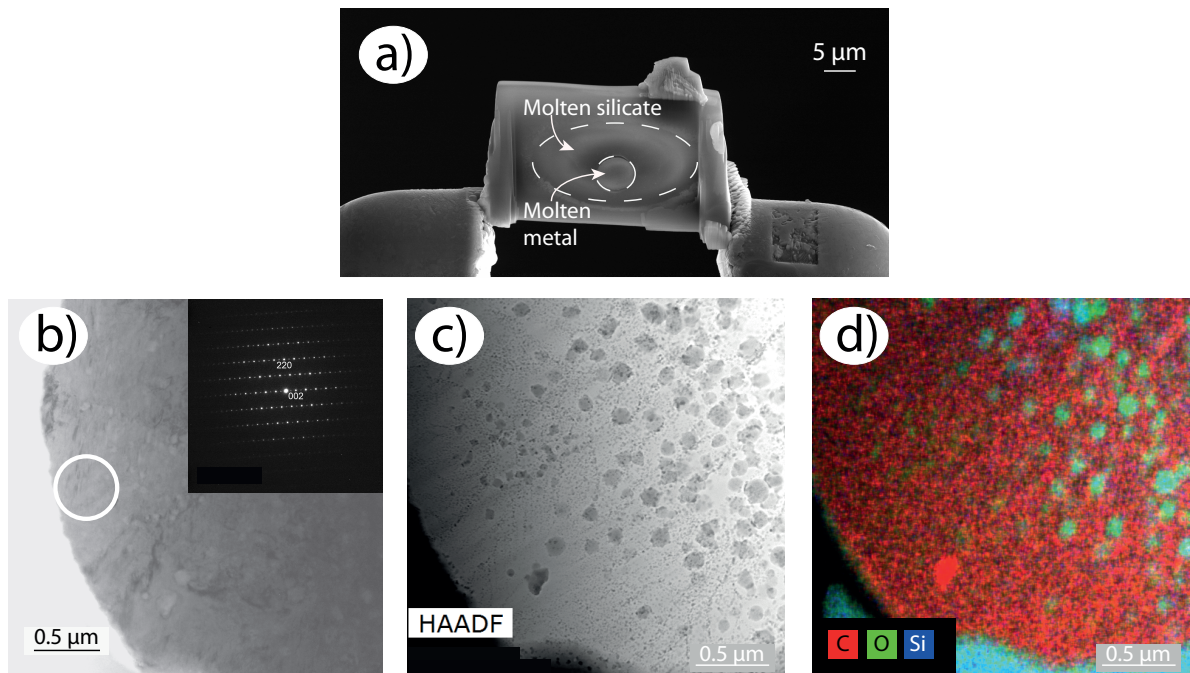
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304   *Figure 1: **Images of run BAS C 39 synthesized at 49 GPa and 3600 K**. a) Secondary electron*

305   *image where two different phases can be observed in the sample: the molten silicate and the*

306   *molten metal that were equilibrated at high pressure and high temperature. b) Bright field TEM*

307   *image and selected area electron diffraction pattern (Inset) of the metallic phase showing the*

308   *presence of stochiometric Fe_7C_3 (also confirmed with EELS). c) High-angle annular dark field*

309   *of the same sample showing exsolutions that developed during quenching along with d)*

310   *Scanning TEM-EDS map of the same area which shows exsolved inclusions that are oxygen*

311   *rich (green) and silicon-rich (blue). The bright red inclusion in image c) is believed to be an*

312   *exsolved diamond[27].*
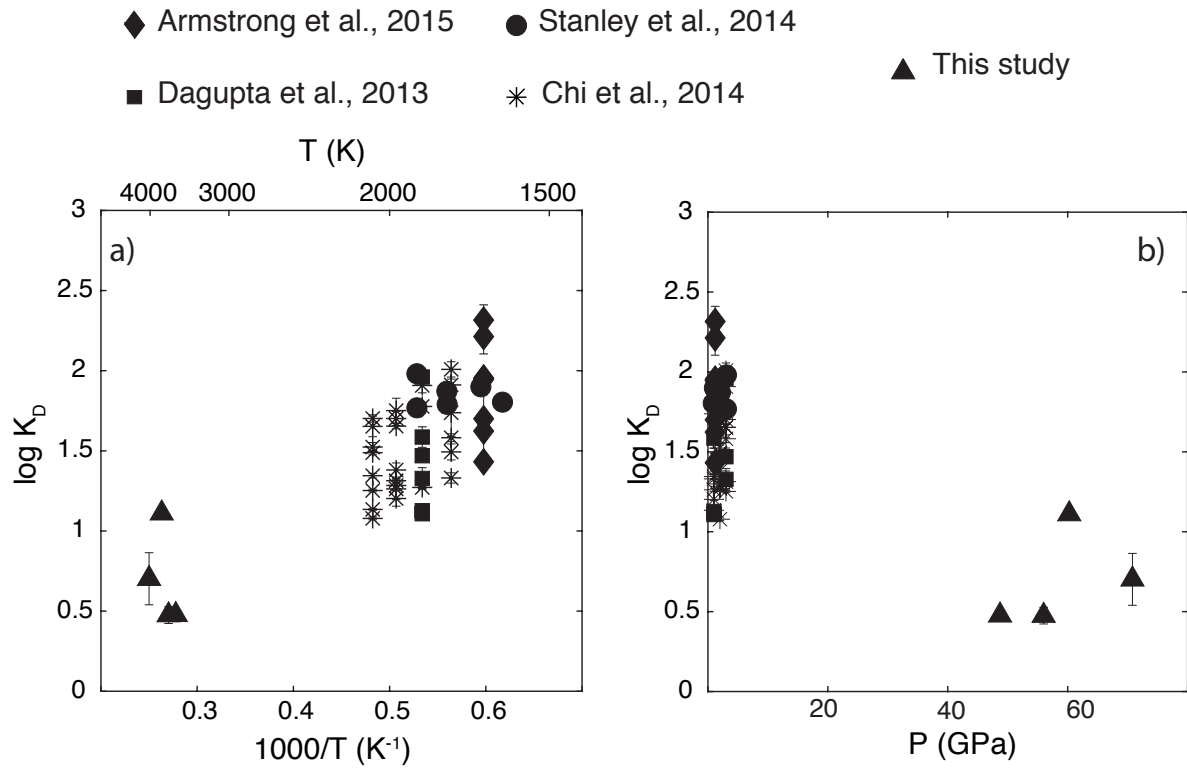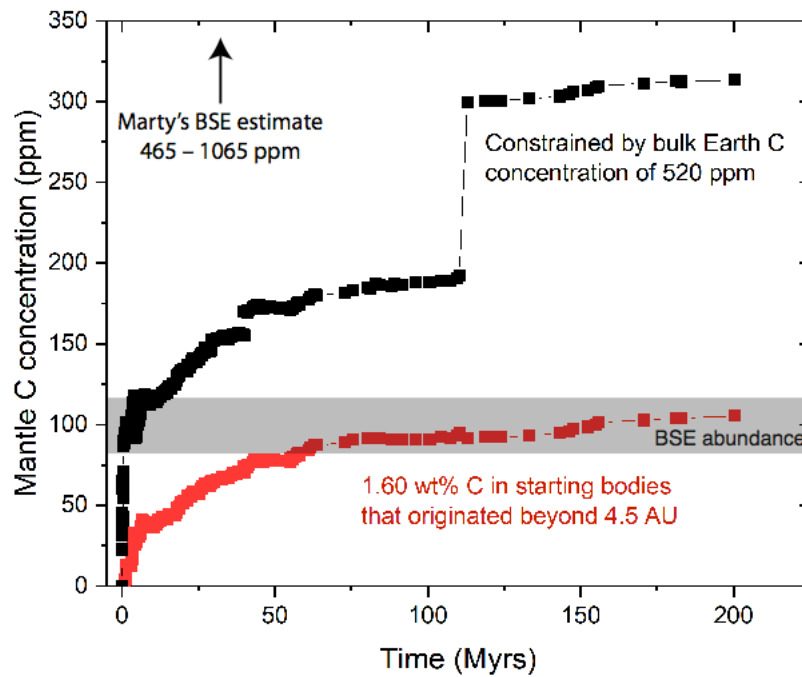
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314 *Figure 2:* **Comparison of our data with results of previous studies with respect to**

315 **temperature (a) and pressure (b).** *Uncertainties on temperature and pressure measurements*

316 *for diamond anvil cell experiments are of 300 K and 5 GPa respectively.*

317

318

*Figure 3: **Mantle C concentration during accretion as a function of time based on a***

***combined accretion/differentiation model**[9,10]. The red symbols show the evolution of mantle*

carbon concentration assuming that all starting planetesimals that originated from beyond

4.5 AU contained an average of 1.6 wt.% C; bodies originating at heliocentric distances <

4.5 AU are assumed to contain zero C. The black symbols show the evolution when the

average carbon concentration in starting bodies originating from beyond 4.5 AU is 1.6 wt.%

and C concentrations in bodies originating at < 4.5 AU decrease along a linear gradient to

reach a value of zero at a heliocentric distance of 1.88 AU. The prominent jump at 110 Myrs

is caused by the final giant impact. Estimates of the BSE concentration range from 100±20

ppm[4], as shown, to 765 ± 300 ppm[20].

Methods

a) Starting materials

329 **Methods**

330 **a) Starting materials**

331 For the silicate starting glass, we mixed oxide and carbonate compounds in stoichiometric

332 proportions, decarbonated over night at 900°C, then added FeO. Powders were ground under

333 acetone for some time to ensure a homogeneous composition. We created pellets that were

334 subsequently fused in argon flux for several seconds at about 1400°C using a levitation furnace

335 device in Orléans, France[39]. Chemical composition of the glass was checked using EPMA

336 ("Basalt" in table S1) and we could verify that no compositional variation was detectable. The

337 glass sphere was subsequently polished down to a thickness of 20 μm and machined in IPGP,

338 France, to obtain small disks of 80 microns diameter[25].

339 The goal of this study is to understand the effect of core formation on the partitioning of carbon

340 between Earth's most important reservoirs: the silicate mantle and the metallic core. Carbon is

341 a prominent contaminant in the laboratory, with multiple sources of contamination, from the

342 use of ethanol and acetone, various type of glue or ambient contamination. It has also been

343 suggested that diamonds used as pressure transmitting tool can also diffuse carbon to the

344 samples[40]. To better understand the origin of carbon, we used ^{13}C as the source of carbon in our

345 experiments. We synthesized the metallic phase using piston-cylinder apparatus. In order to

346 assess the extent of carbon contamination during LH-DAC experiments, we only used ^{13}C. We

347 mixed 5 wt.% of ^{13}C powder (97% pure from Cambridge Isotope Laboratories, Inc.) with 95

348 wt.% of Fe. We melted this alloy at 2 GPa and 1873 K in MgO capsule for 10 minutes using a

349 ½" piston-cylinder assembly at BGI. This ^{13}C-doped carbide was then crushed to be used for

350 the metallic part of the for laser-heated diamond anvil cell experiments. The composition was

351 checked by EPMA (see table S1, "Metal") using an Fe_3C standard (see sections d) and e)).

352

353 **b) Laser-heated diamond anvil cell experiments (LH-DAC)**

354   We used diamonds with 250 μm culets and rhenium gaskets. Gaskets were pre-indented to

355   obtain a thickness of about 40 to 50 μm, and subsequently laser-drilled to have an experimental

356   chamber of about 90 μm in diameter. We then loaded two silicates discs encapsulating a flake

357   of carbide[25]. Samples were compressed to the target pressure, and then laser-heated at the

358   desired temperature using a doubled sided laser heating system at BGI. Temperature was

359   generated by two fiber continuous-wave (CW) YAG lasers (SPi©) with wavelength of 1064

360   nm and delivering 100 watts each. The laser beam was focused onto the sample using two NIR-

361   Mitutoyo objectives lenses with x20 magnification. Temperature was measured simultaneously

362   and continuously on both sides of the diamond cell using a spectro-radiometric technique[41]

363   using a 2500i spectrometer and pixis 400 CCD camera from Princeton-Instrument© from the

364   light collected through both objective lenses. The peak temperature, above the liquidus

365   temperature of the material, was maintained for few tens of seconds, before switching off the

366   electric power of the lasers to quench the experiment. The samples quenched extremely quickly

367   due to the high thermal conductivity of diamonds and the cells were subsequently decompressed

368   slowly over several hours. As observed in previous studies using LH-DAC, the silicate part

369   quenched to a glass, and the Fe-rich metal presents Si- and O-rich inclusions[24,25].

370

371   **c) Sample recovery**

372   We used the Focused Ion Beam facility (FIB, dual beam Scios FEI©) in Bayerisches Geoinstitut

373   (BGI) to recover small lamellae of 3–5 μm thickness, about 40 μm in length and 20 μm high[24,25].

374   In the samples, we observed two quenched phases that had been molten at high pressure and

375   high temperature: a silicate and metallic one (see Fig. 1-a in the main text).

376

377   **d) Fabrication of analytical standards**

378   For EPMA analysis of the carbon contents of metals, we synthesized a Fe_3C (cementite) carbide

379   standard at BGI[42], by inserting a 1 mm diameter, 10 mm long, 99.99 % purity Fe wire in a thick

380   graphite sleeve and heating it to 1423 K at 15 kbar in a piston cylinder for one week. The

381   reaction product was confirmed to have the Fe_3C cementite structure by XRD and was assumed

382   to be stoichiometric.

383   For the nanoSIMS analysis of quenched glasses, we synthesized ^{13}C-doped glass standards[43].

384   We first mixed oxides in basaltic proportions and made a glass at 1600°C in a furnace for two

385   hours. This glass was analyzed by EPMA for major elements, and by FTIR to confirm that it

386   was carbon-free ("B1" in table S1). Subsequently, we performed piston-cylinder experiments

387   to incorporate ^{13}C into the basaltic glass. The source of ^{13}C was chosen as oxalic acid enriched

388   in ^{13}C (Cambridge Isotope Laboratories, Inc.), that was loaded along with the basaltic glass in

389   a Pt capsule and pressurized to 2 GPa and heated to 1600°C for 10 minutes[43]. We created two

390   glass standards (B1145 and B1147) containing 785 ppm and 1263 ppm of carbon respectively,

391   as measured by FTIR at BGI. For the FTIR measurements, we used an extinction coefficient[44]

392   of 69500 $L.mol^{-1}.cm^{-1}$. We report the FTIR measurements along with EPMA analyses on table

393   S1 for the carbon-free (B1) and the two carbon-doped glasses.

394

395   **e) EPMA analysis**

396   Major elements from both the metallic and silicate phases were analyzed by Electron Probe

397   Microanalysis (EPMA). We calibrated using wollastonite, spinel and olivine for Ca, Al and Si,

398   Mg, Fe in the silicate respectively with current of 15 keV and 15 nA, a focused beam and a phi-

399   rho-Z correction. For the metallic phase, we used a pure Fe wire, FeSi, Fe_2O_3 and Fe_3C

400   standards to calibrate for Fe, Si, O and C respectively, at 15 keV with a 25 nA beam current,

401   again using a focused beam and phi-rho-Z correction. The counting time was 10 seconds on the

402 background, and 20 seconds on the peak for all elements, and the samples and Fe₃C standard

403 were not carbon coated.

404 Analyzing carbon using the microprobe requires special consideration[45]. In order to quantify

405 the carbon content of both the experimental metals and the metal starting material, we

406 synthesized a Fe_3C carbide primary standard (see section d)). During C analysis, we took

407 particular care in repeating the measurements on the metallic standards to quantify background

408 levels (contamination) inside the EPMA. We measured the count rate from the C kα peak on a

409 pure Fe standard continuously for ten minutes, and found no statistically-significant change in

410 peak height over that timescale[42]. We performed repeated measurements of pure iron wire to

411 check for carbon contamination, and observed a background for carbon of 0.49±0.07 wt.%

412 which is well below the amount of carbon present in the metallic phase of our samples. The

413 same was done with pure FeSi, for which we measured 0.36±0.05 wt.% of carbon in the

414 standard. As for oxygen contamination, we measured a background of 0.04±0.04 wt.% in the

415 Fe wire and 0.15±0.05 wt.% in FeSi. Those values are again well below the amount measured

416 in our samples. Standard analyses were consistent and stable over the whole duration of the

417 analytical sessions.

418 Analyses performed on the quenched silicate highlighted high concentrations of FeO (from 14

419 to 27 wt.%), as observed previously in similar experiments[24,25]. In the metallic phase, we

420 observe a range of concentrations of carbon, from 5 to 9.5 wt.%, depending on the sample.

421 Oxygen and silicon are also present in the metallic phase, from 2.8 to 7.5 wt.% oxygen and 0.3

422 to 5.6 wt.% silicon. We report on tables S2 and S3 the full analyses of our recovered runs for

423 silicate and metal respectively. Whilst some analytical totals are low, these low totals are not

424 explained by the thin samples, as a 3 μm deep lamella is thick enough to not lose electrons, nor

425 significant X-ray flux, through the base[46].

426

f) nanoSIMS analysis

428 The abundance of carbon in the silicate phase was expected to be extremely small, due to its

429 high siderophility[1]. Laser heated diamond anvil cell experiments produce very small samples,

430 with silicates phases that are only few micrometers wide. Therefore, NanoSIMS was used in

431 this study to achieve required analytical precision and spatial resolution and quantitatively

432 analyze the low carbon contents in the silicate phase.

433 NanoSIMS measurements are sensitive to matrix effects, so we took particular care in

434 synthesizing and using relevant standard glasses (see section d)). Along with those two

435 standards, we used two standards of natural rocks containing natural and known amounts of

436 carbon: a basaltic glass from the East Pacific Rise[47,48] ALV 981-R23 containing 405 ppm of

437 CO_2; and a piece of glass from D'Orbigny meteorite[49,50] containing 40 ppm of C. Composition

438 of those standards and of the natural samples are given in table S1. During the NanoSIMS

439 session, we analyzed the standards each day to check for consistency and demonstrate the

440 reproductivity of the measurements.

441 NanoSIMS analyses were performed using the CAMECA NanoSIMS 50L at the Open

442 University (Milton Keynes, UK). Prior to NanoSIMS analysis, a large area of each sample was

443 pre-sputtered using a focused primary beam of 16 KeV Cs^+ ions with probe current of 100 pA

444 to remove surface contamination. The sizes of the pre-sputtered areas vary from 5×5 to 7×7

445 μm^2. Analyses were then carried out in spot mode first by scanning a 50 pA Cs^+ beam onto the

446 inner 3×3-5×5 μm^2 areas, with secondary ions of $^{12}C^-$, $^{13}C^-$, $^{30}Si^-$, $^{24}Mg^{16}O^-$, $^{27}Al^{16}O^-$ and

447 $^{56}Fe^{16}O^-$ collected in electron multipliers simultaneously. Only the data from the inner 60% of

448 the area were collected using electron gating to avoid carbon contamination from the

449 surrounding areas. Each spot analysis consisted of 200 measurements, with a total analysis

450 time of ~2 minutes.

451 After spot analyses, selected sample areas were also measured in imaging mode. A 50 pA Cs^+

452 beam was also used in imaging mode, with five secondary ions ($^{12}C^-$, $^{13}C^-$, $^{30}Si^-$, $^{56}Fe^{13}C^-$ and

453 $^{56}Fe^{16}O^-$) monitored simultaneously. Each imaging analysis consisted of 10 frames. A frame

454 size of 256×256 pixels was used for all images with an integration time of 500 ms per pixel,

455 leading to total analysis time of 5-6 minutes for each image.

456 The mass resolving power (m/Δm) for both spot and imaging mode was set to 9000 (CAMECA

457 definition), sufficient to resolve all interferences from neighbouring mass peaks, such as ^{13}C

458 from ^{12}CH. We could extract from the region of interest (ROIs) the ratios of $^{13}C/^{30}Si$ and

459 $^{12}C/^{30}Si$. We derived a precise calibration line using those standards, which was used to infer

460 the amount of carbon (both ^{12}C and ^{13}C, see figures S1-a and S1-b) present in each of our

461 samples. During analyses, we carefully avoided metal blebs that are sometimes present in the

462 silicate phase in this style of experiments, interpreting them to be entrained rather than exsolved

463 on quench. We present in Fig. S2 NanoSIMS ion images of the metallic phase of sample BAS-

464 C-39, to highlight the level of detail that can be reached with NanoSIMS.

465 Some uncertainty of the NanoSIMS measurements may derive from the difference between the

466 composition of the samples and the one of the standards. We calculated the amount of carbon

467 in each of our samples using the C/Si ratio of the standards and the experimental runs for both

468 ^{12}C and ^{13}C. The standards we used contained between 40 and 50 wt.% of SiO_2 whereas the

469 runs contained between 35 and 40 wt.% SiO_2. To encompass the range of potential sources of

470 uncertainty, we have assigned an uncertainty of ± 15% to the nanoSIMS measurements.

471 Analysing our samples and standards with the NanoSIMS revealed that the carbon present in

472 both cases was not only ^{13}C but also ^{12}C, with a lower $^{13}C/^{12}C$ ratio in experimental silicates

473 than in the metal (values from 2.8 to 6.9 for metal versus 0.37 to 1.7 for silicate, see Fig. S1-c).

474 The presence of significant concentrations of ^{12}C was unexpected, since we carefully prepared

475 the samples using only ^{13}C, but demonstrates that carbon contamination is probably ubiquitous

476   in high-temperature DAC experiments. The silicate starting powders were heated overnight at

477   900 °C and then converted to glass using an aerodynamic levitation system at temperatures of

478   about 1400 °C. Thus, despite several hours at elevated temperatures, it is extremely hard to

479   eliminate the presence of environmental carbon. ^{12}C could originate from the diamond anvils,

480   the carbonate powders being not fully decarbonated, by sorption of carbon from the atmosphere

481   or from residues of organic carbon contamination during sample preparation.

482   The $^{13}C/^{12}C$ measured in the experimental silicates was lower than in the corresponding metals

483   (Fig. S1-c). This indicates that experiments did not reach isotopic equilibrium, and implies that

484   the most significant source of carbon contamination was the silicate starting material rather than

485   the diamond anvils. This observation is contrary to what has been proposed in previous LH-

486   DAC studies[26,40]. We also measured silicate in unmelted regions in DAC samples BAS C 39

487   and BAS C 42 at the NanoSIMS, that contained 386 and 347 ppm of carbon ($^{12}C + ^{13}C$)

488   respectively, highlighting the presence of carbon in the starting material. We demonstrate here,

489   for the first time with support of NanoSIMS measurements, that carbon contamination in our

490   DAC experiments is not due to carbon diffusion from the diamond anvils to the samples, but is

491   from the samples themselves.

492

493   **h) TEM analysis**

494   We performed transmission electron microscope (TEM) analyses on a FEI Titan G2 80-200

495   S/TEM equipped with X-ray energy dispersive spectrometer (EDS) and electron energy-loss

496   spectrometer (EELS) in Bayerisches Geoinstitut in order to observe and further analyze the

497   experimental metals. To do so, we further thinned down one of our samples to a thickness of

498   60-80 nm using FIB. The FIB lamella was plasma cleaned prior to TEM analysis in order to

499   remove surface contamination. We determined Fe/C ratios in our sample and also perform EDS

500   mapping in the metallic phase. The quantification of the EELS analyses followed the procedure

501 previously described[40,51], using experimentally-determined ratios of partial cross sections of C

502 K and O K edges against the Fe L edge versus sample thickness, which were calibrated with

503 synthetic $Fe_{0.94}O$ and Fe_3C samples. Note that due to the inhomogeneity of the metallic phase

504 of the sample (see Fig. 1-a,-b,-c), EELS technic cannot provide a measurement of the bulk

505 carbon contents of this phase.

506

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543

Extended Data Tables and Figures

545

546 *Table S1: Composition of the synthetic basaltic starting silicate used for the LH-DAC*

547 *experiments ("Basalt"), the standards for the NanoSIMS synthesized with piston-cylinder at 2*

548 *GPa and 1600 ℃ ("B1145" and "B1147"), the carbon-free starting material for piston-*

549 *cylinder experiments ("B1") and the starting metal used in LH-DAC experiments. We also*

550 *report the composition of the two natural samples that were used at the NanoSIMS as*

551 *standards. b.d. stands for below detection limit.*

Samples	Basalt	B1145	B1147	B1	D'Orbigny glass[a]	ALV 981-R23[c]		Metal
wt.%	*N=10*	*N=7*	*N=7*	*N=7*	-	-		*N=14*
SiO_2	52.18 (0.15)	48.05 (0.18)	50.89 (0.27)	53.56 (0.13)	41.7	49.63	C	5.67 (1)
Na_2O	1.90 (0.03)	1.80 (0.06)	1.86 (0.06)	2.03 (0.03)	-	2.91	Fe	91.61 (1.31)
CaO	10.92 (0.07)	9.47 (0.10)	10.03 (0.08)	10.70 (0.05)	23.3	11.61		
FeO	11.35 (0.04)	13.65 (0.20)	10.71 (0.06)	10.04 (0.06)	11.0	8.16		
Al_2O_3	14.66 (0.07)	16.37 (0.11)	15.22 (0.10)	15.48 (0.05)	19.6	16.60		
MgO	7.45 (0.07)	7.22 (0.08)	7.57 (0.10)	7.73 (0.06)	1.87	8.36		
C (ppm)	-	785 (8)*	1263 (13)*	b.d.*	40	111		
Total	98.46 (0.20)	96.66 (0.19)	96.41 (0.33)	99.55 (0.15)	99.3[b]	98.96[d]		97.28 (0.76)

552 *Measured using FTIR (see text for details)
553 [a] Composition from ref.[50]
554 [b] Total includes also 2 wt.% of TiO_2, Cr_2O_3, MnO
555 [c] Composition from refs.[47,48]
556 [d] Total includes TiO_2, MnO, K_2O and P_2O_5

557

558 *Table S2: Compositions of quenched silicate liquid. Major elements were analysed using*

559 *EPMA and carbon was analysed using NanoSIMS. See text for details.*

Run #	BAS C 50	BAS C 60	BAS C 39	BAS C 42
wt.%	*N=4*	*N=5*	*N=8*	*N=6*
SiO_2	35.84 (0.38)	39.97 (1.98)	34.09 (0.83)	39.66 (0.99)
MgO	8.85 (0.20)	8.60 (0.28)	9.53 (0.51)	9.02 (0.26)
Al_2O_3	19.46 (0.12)	25.93 (0.50)	16.34 (1.36)	24.48 (0.64)
CaO	4.02 (0.16)	4.89 (0.12)	5.67 (0.20)	5.68 (0.31)
Na_2O	2.13 (0.07)	2.22 (0.10)	2.32 (0.15)	2.53 (0.11)
FeO	23.82 (1.40)	18.60 (0.60)	27.35 (2.55)	13.71 (1.72)
C (ppm)*	312 (47)	545 (82)	2805 (420)	272 (41)
Total	94.12 (1.85)	100.2 (1.96)	95.30 (2.34)	95.07 (1.55)

560 *15% uncertainties estimated on NanoSIMS measurements (see text for more details).

561

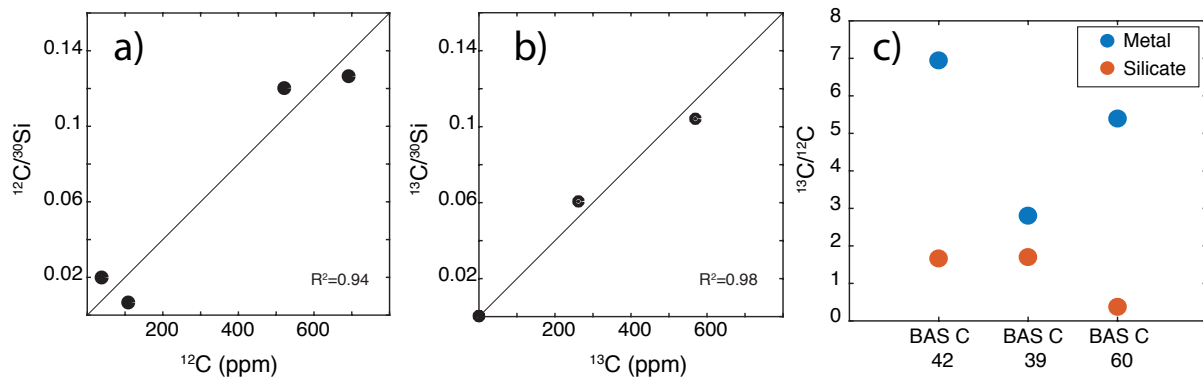562 *Table S3: Composition of the metal phase of our recovered runs analysed using EPMA. See*

563 *text for details.*

Run #	BAS C 50	BAS C 60	BAS C 39	BAS C 42
EPMA (wt.%)	*N=3*	*N=3*	*N=4*	*N=2*
Fe	*81.68 (1.25)*	*79.56 (0.9)*	*85.96 (0.58)*	*78.01 (0.78)*
Si	*0.46 (0.16)*	*5.56 (0.16)*	*0.33 (0.11)*	*2.65 (0.40)*
O	*3.79 (0.89)*	*7.56 (0.37)*	*2.85 (0.48)*	*3.42 (0.10)*
C	*5.90 (0.18)*	*4.99 (0.28)*	*9.44 (0.24)*	*5.35 (0.20)*
Total	*91.83 (0.41)*	*97.66 (0.38)*	*98.57 (0.29)*	*89.43 (0.48)*

564

565
566

567

568 *Figure S1:* **Results from NanoSIMS measurements.** *a) and b) calibration lines obtained for*

569 *^{12}C and ^{13}C respectively using our standards and two natural samples (d'Orbigny and ALV*

570 *981-R23) and both FTIR and NanoSIMS measurements. Note that for ^{13}C measurements, the*

571 *two natural samples plot at 0. c) Illustration of the isotopic disequilibrium between the*

572 *metallic and the silicate phase for three of our recovered runs which illustrates the presence*

573 *of ^{12}C in our reacted samples, despite the use of pure ^{13}C in the starting material.*

574

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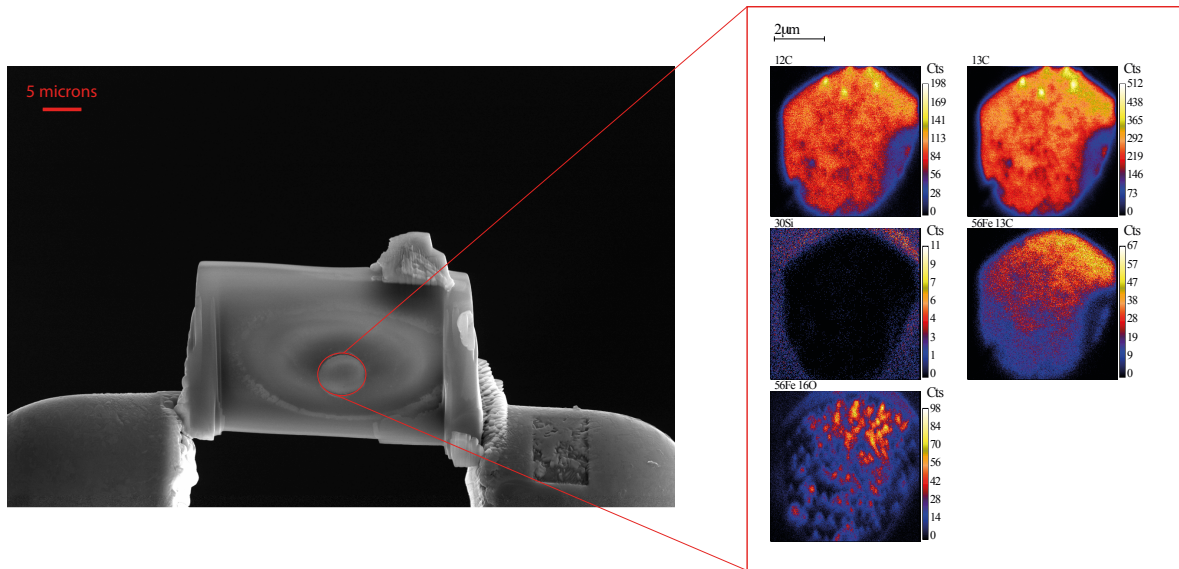
Figure S2: Secondary electron image of sample BAS-C-39 along with NanoSIMS mapping of
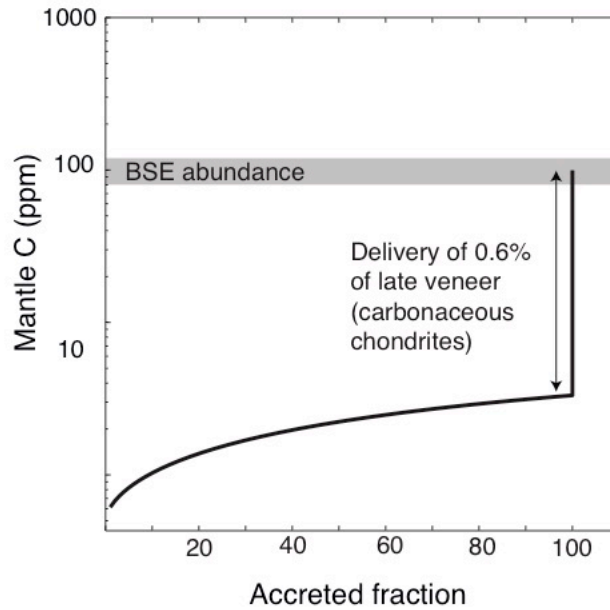
the metallic phase.

578

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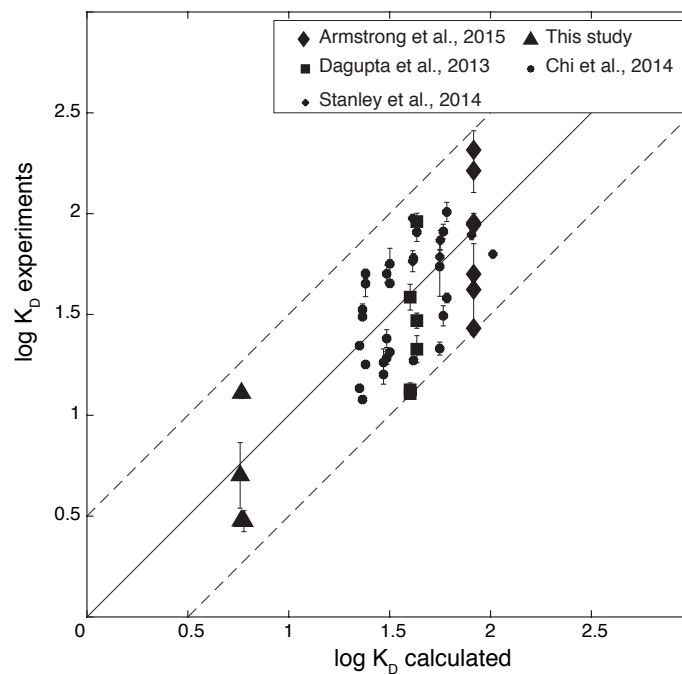
*Figure S3: **Validation of our partitioning model**. Comparison of the K_D values (see eq. 3 in*

the main text) calculated from the experiments and from our model (eq. 4 in the main text).

582

583

Figure S4: **Continuous core formation model**. *Evolution of the concentration of carbon in the mantle during Earth's core–mantle differentiation and the late accretion stage based on a continuous core formation model. The estimated BSE concentration of 80–120 ppm is shown in both cases*[4].

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601

602   **Authors contributions**

603   I.B. conceived the project, performed experiments, analyzed data and derived the

604   thermodynamic model. E.S.J. analyzed data. I.A.F., X.Z. performed NanoSIMS measurements.

605   S.P. performed laser heating experiments and helped with FIB. N.M. performed TEM

606   measurements. D.C.R. and S.A.J. built the accretion model. I.B., E.S.J. and D.C.R wrote the

607   manuscript.

608

609   **Additional information**

610   **Supplementary Information** is available for this paper. Correspondence and requests for

611   materials should be addressed to Ingrid Blanchard (ingrid.blanchard@uni-bayreuth.de).

612

613   **Supplementary Information**

614   Using our high pressure and high temperature experiments along with the ones of the literature,

615   we built a new parameterization for the metal–silicate partitioning of carbon (Eq. 4 in the main

616   text). We compare in Fig. S3 the agreement between the experimental K_D and the one predicted

617   by our regression. Then, we utilized two styles of modelling in this study. The first one is the

618   one presented in the main text, and the second one is a more simple continuous core formation

619   model that has been applied in many geochemical studies[1–3]. Here, Earth is assumed to accrete

620   material in small (1%) increments and each batch of accreted metal is assumed to equilibrate

621   chemically with the entire mantle. The equilibration pressure is assumed to correspond to

622   conditions the base of the magma ocean and increases as the planet grows. The temperature at

623   the base of the magma ocean, which is constrained by the peridotite solidus–liquidus, also

624 increases as the Earth grows. For simplicity, we set the fO$_2$ during accretion at IW-2.3 as given

625 by the current mantle/core FeO/Fe ratio[4].

626 The concentration of carbon in the mantle today is estimated to lie between 80 and 120 ppm

627 (Dasgupta and Hirschman 2010; Hirschman 2016). On the other hand, the bulk Earth carbon

628 content has been proposed to lie between 520 ppm[5] and 750 ppm[6]. Here, we assume that each

629 stepwise-accretion step delivers material with a bulk C content that matches the bulk Earth

630 value[5]. Using the higher value of 750 ppm[6] would not significantly change the results. The

631 partition coefficient of carbon is calculated after each equilibration step following eq. 4, and

632 the resulting amount of carbon present in both mantle and core is derived from this calculation.

633 Hence, carbon is present throughout accretion.

634 The Earth is believed to have been bombarded by bodies comprised of chondritic material after

635 core formation ceased (the so-called late accretion stage which resulted in addition of the "late

636 veneer") in order to explain the concentrations of highly siderophile elements present in the

637 mantle[7–9]. The amount of material that was delivered to Earth during late accretion was about

638 0.6% of Earth's mass[10], and was not incorporated into the core, but rather remained in the

639 mantle. Chondrites are carbon-rich, with up to 3.2 wt.% of carbon reported in CI chondrites[11],

640 so their delivery to Earth brought a substantial amount of carbon. We incorporated the effect of

641 such late accretion in our model by assuming that the carbon content of the late veneer matches

642 the mean carbonaceous chondrite (CI, CM, CO and CV) value of 1.6 wt.%.

643 Results of the continuous core formation model are presented in Fig. S4. The combined effects

644 of core formation and later addition of late veneer are needed in order to reproduce the carbon

645 content of the BSE. Core–mantle differentiation effectively strips the mantle of all carbon and

646 the addition of C during late accretion is required to achieve the mantle carbon concentration.

647 According to this model, the resulting concentration of carbon in the core after the combined

648 effect of differentiation and late veneer is 0.16 wt.%. This result is consistent with previous

suggestions, based on first principles molecular dynamics[12] and experiments[13], that carbon is not a dominant element in Earth's core. As discussed in the main text, this simple scenario is not coherent with astrophysical accretion scenarios. This model assumes that the bulk Earth C-content is known *a priori*, and the assumption that all accreted metal equilibrate with all the mantle is also unrealistic.

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