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1 **Growing Diamonds in the Laboratory to investigate Growth, Dissolution,**
2 **and Inclusions Formation processes**

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26 **ABSTRACT**

27

28 Replicating lithospheric diamonds experimentally helps reveal their histories at depth, which
29 may be complicated by successive growth and dissolution episodes. Here, we present
30 diamond growth and dissolution experiments to constrain the conditions of diamond
31 formation and residence in the Earth's lithosphere before their transport to the surface.
32 Experiments were performed on mixtures of carbonates, natural lherzolite or MORB, water,
33 diamond seeds, and with or without graphite using multi-anvil presses over a few hours to
34 more than a day at conditions relevant to the lithosphere (7 GPa, 1300–1670 °C). We
35 observed growth within a volatile-rich melt resulting from the miscibility of silicate-carbonate
36 melt and aqueous fluid via the reaction: Carbonate minerals + Silicate minerals/glasses + H₂O
37 = [volatile-rich melt] ± minerals + C_{diamond} + O₂. In the absence of graphite, we observed
38 diamond dissolution within the same hydrous-carbonate-silicate-bearing melts and under
39 similar pressure and temperature conditions used to grow diamond, indicating that diamond
40 formation requires an oxygen sink (graphite, metallic and/or sulphide melts). In dissolution
41 experiments, we also observed resorption features similar to those described in lithospheric
42 monocrystalline diamonds, which we thus attribute to mantle fluids and not to kimberlite-
43 induced resorption during magma ascent. We show that dissolution and growth may alternate
44 in the mantle, and that protracted periods are not necessary to explain natural diamond
45 histories. During experimental growth, inclusions were trapped in diamonds as they are in
46 nature. We observed both syngenetic (formed during growth, representing the parental fluid)
47 and protogenetic inclusions (here particles from the capsule), indicating that both kinds of
48 inclusions can be trapped within a single growth event. Our experiments confirm that
49 inclusions trapped in natural diamonds do not necessarily represent remnants of their parental
50 fluids and are not necessarily contemporaneous: two inclusions near each other within a single
51 monocrystalline diamond may have different histories, and inclusions must be shown to have
52 achieved chemical and isotopic equilibrium before being considered synchronous.

53

54 **1.Introduction**

55

56 In 1964, Harrison and Tolansky described the complex structure of an octahedral diamond,
57 which indicates that significant dissolution and regrowth mechanisms occurred successively, a

58 process since observed in many diamonds (e.g., Smit et al., 2016; Gress et al., 2018;
59 Fedortchouk, 2019; Harris et al., 2022).

60 In the first lines of their 1964 article, Harrison and Tolansky also asked key questions about
61 diamond growth: whether they grow from solution or from a melt, and whether they represent
62 transformed graphite. Although diamonds most likely grow from fluids, and in the following
63 we will call a fluid “any phase that can flow”, through a metasomatic process (Navon et al.,
64 1988), questions remain about the nature of the fluids: melt (dry or volatile-rich), aqueous
65 solutions, supercritical fluids. Numerous attempts have been made to characterize such mantle
66 fluids by studying fluid inclusions trapped in natural diamonds (e.g., Navon et al., 1988;
67 Weiss et al., 2009, 2015, 2022; Nimis et al., 2016; Klein-BenDavid et al., 2004, 2010; Jablon
68 and Navon, 2016), by growing diamonds in the laboratory at mantle pressure (P) and
69 temperature (T) conditions (Pal’yanov et al., 1999, 2013, 2022; Sokol and Pal’yanov, 2008;
70 Pal’yanov and Sokol, 2009; Bureau et al., 2012, 2016, 2018), or by modelling diamond
71 growth from fluids (e.g., Sverjensky et al., 2015; Huang and Sverjensky, 2020; Mikhail et al.,
72 2021; Rinaldi et al., 2023). It appears that worldwide lithospheric diamonds, whether
73 monocrystalline, fibrous, coated (monocrystalline core and fibrous rim), or polycrystalline,
74 grow from CHO-rich supercritical fluids comprising silicate, water, carbonate, and halogens
75 (Jacob et al., 2014; Bureau et al., 2016; Jablon and Navon, 2016). However, depending on the
76 context (i.e., peridotitic mantle or subduction zones, depth in the mantle, and redox
77 conditions), fluids may differ in their nature and composition (Sokol et al., 2009, 2017;
78 Pal’yanov et al., 2013; Stachel et al., 2017) between endmembers, whether aqueous (possibly
79 saline) or melt (carbonate or silicate) in the case of lithospheric diamonds. The parental fluids
80 may vary within a single diamond (Weiss et al., 2022). Among several examples, fibrous
81 diamonds from Diavik exhibit compositional and isotopic ranges of fluid inclusion
82 compositions (assumed to represent the parental fluid) in concentric layers, evidencing their
83 complex growth from successive batches of fluids (Klein-BenDavid et al., 2004).
84 Furthermore, isotopic profiles (i.e. $\delta^{13}\text{C}$, $\delta^{15}\text{N}$) are common within single diamonds (e.g.,
85 Boyd et al., 1994), indicating that diamond growth can occur in several stages.

86 Resorption features have been described in most natural monocrystalline and fibrous
87 diamonds based on their surface morphologies (Fedortchouk, 2019; Harris et al., 2022),
88 cathodoluminescence (CL) imaging (Robinson, 1979; Smit et al., 2016; Smart et al., 2016;
89 Gress et al., 2018; Fedortchouk, 2019), and isotopic profiles. Resorption was long believed to
90 result from interactions between the host diamond and kimberlitic or lamproitic melts during

91 magma ascent (Robinson, 1979). However, based on the observed range of resorption
92 morphologies, it was recently shown that if dissolution occurs in kimberlite magmas
93 (Fetdorchouk et al., 2017), it can also occur in the mantle at depth in association with
94 metasomatic events (Gurney et al., 2004; Fetdorchouk et al., 2019). This assumption was
95 confirmed by high pressure and high temperature experiments mimicking diamond resorption
96 in mantle fluids and devoted to decrypting diamond histories from their surface features (e.g.,
97 Khokhryakov and Pal'yanov, 2010, 2015; Fetdorchouk et al., 2019).

98 Monocrystalline diamonds worldwide contain rare silicate mineral inclusions, i.e., true
99 encapsulated pockets of the mantle. However, the significance of these inclusions may differ:
100 do they form contemporaneously with the diamonds and from the same parental fluid
101 (syngenetic) or are they pre-existing, becoming passively trapped (protogenetic)? In some
102 diamonds, inclusions of sulphides, olivines, pyroxenes, and garnets were shown to be
103 protogenetic (Thomassot et al., 2009; Nestola et al., 2017, 2019) based on either stable isotope
104 geochemistry or comparison of the crystallographic orientations of the inclusions relative to
105 those of their host diamonds. It was proposed that included immiscible sulphide phases
106 become isolated from further isotopic exchange upon encapsulation (e.g. Pearson et al., 1998),
107 thus recording the time of their encapsulation (termed later “synchronous”; Nestola et al.,
108 2017). Although the utility of mineral inclusions for dating diamond crystallization is still
109 being investigated, several individual growth-events were identified from populations of
110 inclusions trapped within a single diamond (Koornneef et al., 2017; Gress et al., 2021); these
111 events may have been separated by billions of years, although their duration and frequency
112 remain unknown. The challenges of determining the time of diamond crystallization were
113 well detailed in a recent review (Smit et al., 2022).

114 To fully utilize the information recorded in diamonds, three processes must be entirely
115 understood: (1) diamond growth in the lithosphere (and in the transition zone and lower
116 mantle); (2) diamond dissolution and the duality between dissolution and growth (i.e., when
117 and how both processes operate in the mantle); and (3) the trapping of silicate and fluid
118 inclusions in diamonds.

119 Replicating diamonds with inclusions similar to those in natural diamonds is a reliable tool to
120 better understand diamond formation processes.. Here, we grew diamonds in natural
121 lithospheric mantle melts, trapping inclusions of various natures. We show that diamonds may
122 grow and dissolve within the same type of parental fluid over very short times. We suggest

123 that several successive processes during diamond's histories at depth may make it very
124 difficult to interpret observations of natural diamonds.

125

126 **2. Materials and methods**

127 *2.1 Starting materials*

128 Powders and glasses of different compositions were employed as starting materials (Table 1):
129 a silicate endmember, carbon sources, diamond seeds, and pure water. For the silicate
130 endmembers, we used two natural powders and one glass. The powders were ground from a
131 natural mid-ocean ridge basalt (MORB) from the Indian Ocean and a natural lherzolite from
132 the Lherz massif (Lorand et al., 2008). The glass was prepared using a laser levitation system
133 to obtain a glass of peridotitic composition and free of crystal nuclei. A portion of the natural
134 lherzolite powder was compacted using a hydraulic press, and the resulting pellet was cut into
135 millimetre-sized pieces that were then levitated in a mixed Argon + H₂ gas flux and melted
136 with an Argon laser at 2500–3000 °C for a few seconds; when the laser was stopped, the
137 levitated silicate melt sphere instantaneously quenched into a glassy sphere. Carbonate
138 endmembers were prepared as mixtures of high-purity synthetic CaCO₃, Na₂CO₃ and K₂CO₃
139 powders, and graphite was added to growth experiments (Sigma-Aldrich®). We oxidized
140 commercial diamond seeds (Microdiamant® synthetic monocrystalline 40–60 µm powder) in
141 a furnace at 1000 °C for 10 minutes to form cavities in their surfaces to favour the formation
142 of inclusions. Furthermore, we added 200-500 µm cubic pieces of broken, gem-quality,
143 monocrystalline type-Ia diamond anvil diamonds (Almax-EasyLab®) to some experiments
144 (Fig. 1A). We loaded roughly equal weights of carbonate, water, and either lherzolite powder,
145 a lherzolite glass sphere (half of the growth experiments, Fig. 1C), or MORB powder (two
146 dissolution experiments) into Pt capsules, as well as various amounts of graphite (growth
147 experiments) and diamond seeds (Fig. 1B) or broken pieces (Fig. 1A); for each experiment,
148 all components were carefully weighed and are presented in the Supplementary Table S1. The
149 capsules were carefully sealed by arc welding to avoid any fluid loss, and after each run, the
150 presence of fluid was checked when opening the capsule. All experiments were saturated with
151 respect to carbon, as indicated by the presence of carbonates and graphite (as round globules
152 or bulk masses, only in growth experiments) in the reaction products.

153

154

2.2 High-pressure experiments

155 Experiments were performed in multi-anvil presses at Bayerisches Geoinstitut, Bayreuth,
156 Germany, and at Laboratoire Magmas et Volcans, Clermont-Ferrand, France, as detailed in
157 our previous studies (see Bureau et al., 2012) and summarized here. The high-pressure
158 assembly comprised a 18-mm edge-length MgO octahedron, in which capsules ($2 \times 2 \text{ mm}^2$ in
159 size) were introduced, together with tungsten carbide anvils with 11-mm edge truncations
160 (Keppler and Frost, 2005). Graphite heaters were used and temperatures were measured using
161 W(3%Re)–W(25%Re) thermocouples. The experiments were performed at 7 GPa following
162 the calibration curves of the presses, at 1350–1670 °C, and run durations ranging from 4 to 27
163 hours. We selected temperatures above those expected for lithospheric diamond growth
164 ($\geq 1100 \text{ °C}$) to accelerate experimental processes because of limited access to the multi-anvil
165 presses. We showed that growth kinetics are slower at lower temperatures. In an earlier study,
166 one similar experiment was performed at 1200°C for 7 days, diamond growth on seeds was
167 observed with SEM (Bureau et al., 2012). *It shows that slightly increasing the temperature of*
168 *100-150°C does not modify the growth process but increases its kinetics..* After quenching to
169 room temperature, the samples were recovered after slow decompression. Capsules were
170 weighed before and after opening to check for the presence of aqueous fluids, and were
171 opened carefully (Fig. 1D) to observe if water leaked out.

172

173

2.3 Focused ion beam milling and scanning electron microscopy

174 The quenched solid products were carefully peaked and gently deposited on stubs covered
175 with carbon tape and studied by Scanning Electron Microscopy (SEM) using a Zeiss
176 Crossbeam Neon40 at Institut de Minéralogie, de Physique des Matériaux et de Cosmochimie
177 (IMPMC, Paris, France). Due to the small sizes of the seed crystals ($< 50 \text{ }\mu\text{m}$), they could not
178 be separated from the surrounding solid matrix. The interiors of some diamond seeds were
179 exposed by Focus Ion Beam (FIB) milling using a gallium beam on a Zeiss Crossbeam
180 Neon40 at IMPMC. When an exposed seed exhibited an inclusion, it was cut on the other face
181 to obtain a thin section (3–5 μm thick) that was then deposited on a silicon wafer for further
182 analysis (Bureau et al., 2018).

183 Quantitative chemical analyses of the samples were performed by energy-dispersive X-ray
184 spectroscopy (EDX) in conjunction with either SEM or FIB. The detector was calibrated with
185 a pure copper target at the beginning of each session to determine the beam current. EDX

186 analyses were calibrated using international ASTIMEX® standards for spectral deconvolution
187 and quantification. The resulting precision on the chemical analyses was about 1 wt.%. Strong
188 carbon contributions from the host diamonds, especially in the case of inclusions, were
189 difficult to avoid and quantify.

190

191 **3. Results**

192 *3.1 Diamond Growth experiments*

193 Our first set of experiments (#50–55; 7 GPa, 1400–1673 °C, 6–24 hours) was designed to
194 grow diamonds in complex fluids (Fig. 2, Table 2a). We used natural lherzolite (powder or
195 glass) as the silicate endmember, synthetic carbonates, synthetic graphite, diamond seeds
196 (powder and/or type-Ia diamond shards), and pure water, with equivalent proportions of fluid
197 (water) and melt (silicates and carbonates). The starting mixtures were inspired by the
198 compositions of micro-inclusions trapped in fibrous diamonds (metal oxides, water,
199 carbonate; Navon et al., 1988) and have been successfully applied in our previous studies
200 (Bureau et al., 2012, 2016, 2018), in which we showed that all diamonds (i.e.,
201 monocrystalline, fibrous, polycrystalline) can grow from these mixtures. We also included
202 experiment #210 from Bureau et al. (2016), in which the silicate endmember was a basaltic
203 MORB glass from the Indian ridge and the carbonate was a natural CaCO₃ from the Indian
204 ridge seafloor (7 GPa, 1350 °C, 6 hours).

205 Once quenched, the capsules were pierced, checked for water leaks, and the solid products
206 were deposited on stubs covered with carbon tape. Samples included carbonates, pieces of
207 glasses, graphite agglomerates, and seeds. In all experiments and at all temperatures, we
208 observed diamond growth in the presence of water, either on seeds (Fig. 2A, B, D, F) or
209 spontaneously (Fig. 2C, E). Further images and descriptions of growths are presented in
210 Figures S1 to S5. Growth was more significant in longer duration runs or at higher
211 temperatures (>1600 °C), in agreement with our previous studies (Bureau et al., 2012, 2016).
212 Indeed, massive growth was observed in experiment #52 (Fig. 2B; performed at 1670 °C for 6
213 hours), in which spontaneously grown diamonds attached to the starting seed, which was also
214 growing (Fig 2B). In experiment #53 (1400 °C, 24 hours; Fig. 2D), large rims grew on the
215 starting cubes, explaining the cubic shape. Growth on cubes was also observed in sample #55
216 (1400 °C, 6 hours; Fig. 2F). Spontaneous growth in the matrix was observed as small
217 octahedral diamonds that grew to up to 8 μm during 6 hours at 1670 °C (Fig. S5).

218 The nature of the carbonate in the lherzolite (K_2CO_3 in run #51; Na_2CO_3 in runs #50 and #55;
219 $CaCO_3$ in run #52; mixtures of all three in runs #53 and #54) and MORB experiments (natural
220 calcite in run #210) did not significantly impact diamond growth, which was of the same
221 magnitude at equivalent pressures, temperatures, and durations (Fig. 2). Compared with
222 previous studies employing different starting silicate materials (synthetic Fe-free, Mg-rich
223 silicate, natural MORB; Bureau et al., 2012, 2016), our experiments confirm that the
224 composition of the silicate does not affect diamond growth.

225 As already mentioned, we only studied the solid portions of the samples. The solid matrix
226 associated with the diamond seeds (or cubic anvil pieces) mostly comprised chemically
227 heterogeneous materials (Table 3), and no silicate crystals were observed, only silicate
228 glasses. The silicate glasses exhibit a large range of carbon concentrations (Fig. 3A, B, Table
229 3, sample #52)) and were associated with tiny carbonate crystals of various compositions
230 depending on the starting carbonates. Carbonate crystals were either free in the matrix or
231 attached to all the diamond seeds (Fig. 2, agglomerated in clusters in Figs. 2C and 3A, B).
232 Small diamonds nucleated within the fluid/melt during the experiments (Fig. 2C, E). In
233 contrast with our previous experiments on other chemical compositions (synthetic, MORB;
234 Bureau et al., 2012, 2016), which produced silicate minerals in addition to glasses and
235 carbonates, the lack of silicates in our present experiments suggests that the experiments were
236 above the liquidus. We do not mean that we are working at conditions where the entire mantle
237 assemblage would be melted, but we propose that melting occurs only locally, where the
238 discrete volumes of metasomatic volatile-rich fluids percolate the solid “dry” mantle and
239 gradually dissolve some minerals from the mantle assemblage around.

240 In sample #52 (7 GPa, 1670 °C), we observed large areas of highly vesiculated glass with a
241 vermicular texture indicating either the presence of coexisting aqueous fluid (lost upon
242 opening the capsule) and volatile-rich melt at the conditions of the experiment (Fig S1),) or
243 the immiscibility process of a unique very hydrous-rich melt due to quench effect. In one such
244 area, we observed octahedral nano-diamonds ($<0.2 \mu m$) *in situ* within the quenched silicate
245 melt (Fig. 3A, B, S1).

246 EDX analyses evidenced strong carbon contamination in the matrix (Table 3; all carbon
247 concentrations expressed as wt.% CO_2). Iron was not present in the matrix because possibly
248 lost to the Pt capsules during the runs (Grove, 1981) and/or partly dissolved in the aqueous
249 fluid phase. The compositions of the amorphous glassy phases associated with diamond seeds
250 were mixtures of the starting silicate and carbonate compositions (Table 3 and Table S2).

251 Some diamond seeds exhibiting significant growth were milled with a FIB, and all were found
252 to contain inclusions. Figure 4 exhibits the four milled diamonds (#53-1, #53-6, #52-5, #52-9)
253 that showed the most significant growth. Inclusions were observed to be isolated or along the
254 initial surfaces of the starting seeds (particularly in diamond #53-1; Fig. 4B, Fig S2). We
255 observed inclusions of different types: mixed glasses with vesicles, fluids (i.e., holes
256 corresponding to fluids lost during FIB milling), and massive metal inclusions. No inclusions
257 contained silicate minerals, whether in experiments with lherzolite powder (starting crystals
258 present) or lherzolite glass (starting crystals absent), confirming that the system was above the
259 liquidus. The absence of crystals can also be explained by the rapid quench following
260 equilibration at high pressure (HP) and high temperature (HT). In nature, though, trapped
261 melts in diamonds do crystallize after their emplacement.

262 The compositions of the glassy inclusions (red crosses in Fig. 4C, F, I, L) are reported in
263 Table 4 (on a volatile-free basis); they were similar to the glassy matrix embedding the
264 diamonds and thus represent the parental fluid. The large metal inclusions were pure platinum
265 (see Fig. 4C and red arrows in Fig. 4B) and were observed together with the glassy/fluid
266 inclusions in several samples, as in our previous studies (Bureau et al., 2012, 2016). These Pt
267 particles were probably either produced as spatter when welding the capsules, or torn from the
268 capsule walls at HP-HT run and trapped as inclusions.

269 The uniform growth observed around the open seed (FIB milling), presented in Figure 4A
270 recalls coated diamonds with monocrystalline cores surrounded by fibrous rims, but here,
271 inclusions were trapped only at the starting surface of the seed and the overgrowth was
272 monocrystalline. Because this diamond was cut perpendicularly to the growth direction, we
273 were able to estimate that it grew 3–5 μm in 6 hours at 1670 $^{\circ}\text{C}$, corresponding to a growth
274 rate of 20 μm per day.

275

276 *3.2 Diamond Dissolution Experiments*

277 Our second set of experiments (#31, #32, #35, #38, #44, #45, #49; 7 GPa, 1300–1450 $^{\circ}\text{C}$, 5–
278 27 hours), was designed to study diamond dissolution (Fig. 5, Fig. S6 and S7, Table 2b). We
279 used either natural lherzolite (#31, #32, #44, #45, #49) or MORB powders (#35, #38) as the
280 silicate endmembers, together with synthetic carbonates, diamond seeds, pure water, and, in
281 experiments #35, #38, and #49, type-Ia diamond pieces. Graphite was not added to the
282 starting materials.

283 Growth did not occur in any of these experiments and diamonds exhibited dissolution
284 features, the most representative of which are shown in Figures 5, S6, S7. Resorptions were
285 more or less pronounced depending on the experimental duration. Matrix products were
286 quenched glasses and carbonates, as observed in our growth experiments (see previous
287 subsection).

288 There were no clear textural differences in seed resorption features between experiments
289 using MORB or lherzolite, suggesting that dissolution did not depend on the composition of
290 the silicate endmember. In our experiments, seeds were partly dissolved (#45, Fig. 5G, Fig
291 S6), exhibit (111) preserved faces (Fig S7), strong trigonal etches at 1350 °C (#44, Fig. 5H)
292 for starting diamond seeds, and etches were cuboctahedral at 1400 °C for starting diamond
293 cubes (#49, Fig. 5I). Resorption was so pronounced that it reached lamination in experiments
294 at 1450 °C (#32, 6 hours, Fig. 5A; #35, 5 hours, Fig. 5B) and graphitization of the seeds after
295 27 hours at 1300–1400 °C (#38, Fig. 5D).

296 Large type-Ia diamond pieces (200–500 μm) introduced with the small seeds in experiment
297 #38 also exhibited significant resorption textures (Fig. 5C, E, F); for example, a 200 μm cube
298 was strongly rounded (Fig. 5C). Some diamonds exhibited crystal rounding and stepped
299 surfaces (Fig. 5E) that could have formed either during growth (Sunagawa, 1984) or
300 resorption (Viljoen et al., 1992). We also observed deeply etched cubic pits (Fig. 5F) in large
301 cubes.

302 In summary, in short-duration experiments, the starting seeds presented trigonal etches and
303 some were laminated, whereas those in the long-duration experiment were transformed into
304 graphite. Large diamond pieces loaded in the short-duration experiments presented
305 cuboctahedral dissolution features, whereas those in the long-duration experiment exhibited a
306 rounded final shape and large cubic pits, with surfaces so etched that they became stepped.

307

308 **4. Discussion**

309 *4.1 Diamond growth versus dissolution in the mantle*

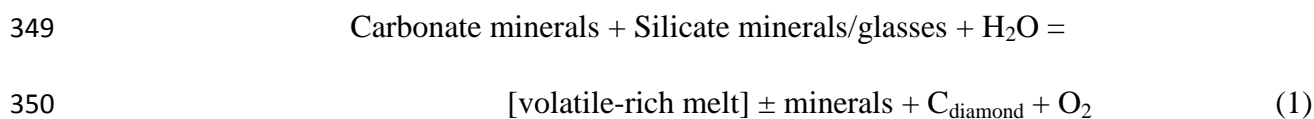
310 Diamond growth occurred in all growth experiments, which contained lherzolite + carbonate
311 (Ca, Na, K, or a mixture) + water + graphite, as is the case when the silicate is iron-free
312 (Bureau et al., 2012) or MORB (Bureau et al., 2016). At the highest temperatures (1670 °C,
313 experiments #52 and #50), a single fluid must be present; it may be either a supercritical fluid

314 or a hydrous super-solidus melt. This was evidenced by the vermicular nature of the glassy
315 quench phase (Fig. 3), suggesting that immiscibility or unmixing due to strong water
316 degassing developed, as observed *in situ* in diamond anvil cell experiments (Bureau and
317 Keppler, 1999), where aqueous fluid and hydrous melt are formed during quenching.
318 Furthermore, inclusions trapped in the diamonds during growth (Fig. 4) contained both glass
319 and void space, which was originally filled by an aqueous fluid before the inclusion was
320 intersected during milling. In run #52 (1670 °C, 7 GPa), nano-diamonds embedded in the
321 glassy layer (Fig. 3) indicate that diamonds nucleated within a hydrous melt. At lower
322 temperatures around 1400 °C, two fluid phases may have been present: a silicate/carbonate
323 melt and a (solute-rich) aqueous fluid. These results are in good agreement with phase
324 diagrams (Wyllie and Ryabchikov, 2000; Dvir and Kessel 2017) showing that at our higher-
325 temperature conditions (7 GPa, 1430–1670 °C), the system lherzolite-CO₂-H₂O is above both
326 the solidus and possibly a critical point. According to Wyllie and Ryabchikov (2000), the
327 minimum depth of the critical point can be constrained from diamonds to 125 km, but there is
328 no constraint on the maximum depth. At depths beyond the critical point, solid peridotite co-
329 exists with supercritical fluids varying continuously in composition (Fig. 6, Tables 3 and 4),
330 in agreement with natural observations (e.g., Klein-BenDavid et al., 2004; Weiss et al., 2015).

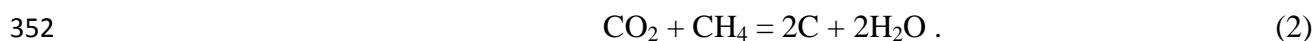
331 Figure 6 shows that part of the compositions of inclusions and matrix glasses in our
332 experiments mirrors that of inclusions trapped in fibrous diamonds, making a trend between
333 silicic endmember and low and high-Mg endmembers (see Fig. S8). We propose that they
334 reflect the discrete metasomatic fluids responsible for diamond growth in the mantle. The
335 range observed between the 3 endmembers recalls recent studies (Mikhail et al., 2021; Rinaldi
336 et al., 2023) proposing that a single fluid may be parent for the formation of eclogitic,
337 websteritic and peridotitic silicate inclusions. The results are consistent with Weiss et al.
338 (2022), who proposed that carbonate-bearing high-density fluids (HDFs) best represent the
339 oxidation state of carbon in diamond-forming fluids, not only for fibrous diamonds, but also
340 for most natural (Jablon and Navon, 2016) monocrystalline diamonds.

341 The oxidation state of the lithosphere is below the Enstatite + Magnesite = Olivine +
342 Diamond buffer (EMOD), meaning that carbonate-bearing fluids/melts or HDFs would
343 unlikely be the source of most diamonds. Nonetheless, Stagno et al. (2013) showed that pure
344 carbonate melt is stable at more reducing conditions than EMOD. Stachel and Luth (2015)
345 also argued that the buffering capacity of the lithosphere is small, and that an oxidized melt
346 (i.e. HDF) can migrate through and react with the reduced lithosphere to form diamonds.

347 Two major reactions that are applicable to nature could describe diamond growth in our
348 experiments:



351 or:



353 Reaction (1) would involve the reduction of either CO₂ or carbonate, and diamond growth
354 would be associated with oxygen consumption. In contrast, reaction (2) is based on carbon
355 oversaturation, and would imply isochemical diamond precipitation and the conservation of
356 oxygen (Deines, 1980; Stachel et al., 2017).

357 If we assume reaction (2), the oxygen fugacity in the lithospheric mantle would be buffered
358 by the fluid rather than the wall rock. In this scenario, diamond precipitation would occur in
359 the presence of two dominant carbon fluid species (CO₂ and CH₄) co-existing with a water-
360 rich fluid with maximal water content (i.e. almost no methane at EMOD; Stachel et al., 2017).
361 This process has been used to model carbon isotopic fractionation during diamond growth
362 from mixed CH₄- and CO₂-bearing fluids, which generates different results relative to
363 diamond crystallization from a single carbon fluid species. It was successfully applied to
364 match *in situ* observed co-variations of δ¹³C, δ¹⁵N, and N content in peridotitic diamonds
365 from Marange, Zimbabwe (Smit et al., 2016), where CH₄ fluid inclusions were found (Stachel
366 et al., 2017). However, this reaction does not explain why diamonds do not grow when
367 graphite is absent, but instead dissolve.

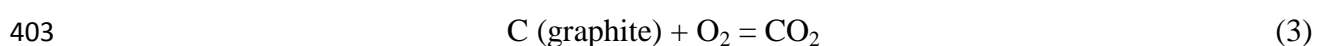
368 If we assume reaction (1) (see Bureau et al., 2016), the volatile-rich melt contains C-O-H
369 species, dissolved in silicate melts (H₂O, OH⁻, and CO₃²⁻ bonded with cations). This reaction
370 applies for all starting silicate compositions (e.g., lherzolite, MORB, synthetic) and
371 carbonates (Na₂CO₃, K₂CO₃, CaCO₃). Carbon can be dissolved in in the hydrous melt, as
372 carbonate groups. In this reaction, the oxygen fugacity would be close to or below EMOD
373 (e.g., Stagno and Frost, 2010). Reaction (1) is therefore consistent with our experimental
374 results.

375 In previous experiments performed at the same pressure (7 GPa) and using ¹³C doped starting
376 carbonates and graphite, we used the isotopic composition of diamonds to estimate the
377 isotopic signature of their source and identify it (Bureau et al., 2018). In that study, a few

378 seeds were milled using a FIB to expose the portions of diamonds that grew during the
379 experiments and trapped inclusions. Among those inclusions, we observed micro-diamonds
380 spontaneously grown in the fluid, then trapped in the seeds within the first few hours of the
381 experiments. We analysed the $\delta^{13}\text{C}$ signatures of those micrometric diamond inclusions by
382 nanoSIMS and compared them with the signatures of the starting materials. The diamonds
383 grown in those experiments had similar signatures ($\delta^{13}\text{C} = -14.19 \pm 3.1\text{‰}$) to those of the
384 starting carbonates ($\delta^{13}\text{C} = -11.40 \pm 0.1\text{‰}$), evidencing growth at isotopic equilibrium with a
385 carbonate source. Furthermore, the isotopic signatures of those diamonds (-14.19‰) could
386 not have resulted from contamination by the starting diamond seeds because the seeds had
387 isotopic compositions ranging from -30.4 to -23.6‰ ($\pm 1.6\text{‰}$). The signatures of the diamond
388 inclusions trapped in new growths also differed from those of the starting graphite ($\delta^{13}\text{C} =$
389 $-26.35 \pm 0.1\text{‰}$), except for diamonds grown at $>1600\text{ °C}$, which had $\delta^{13}\text{C} = -27\text{‰}$ on
390 average, compatible with graphite dissolution in the fluid. That study demonstrated that below
391 1600 °C , only carbonates, and not graphite, are dissolved in the fluid, consistent with the
392 recovery of graphite agglomerates after low-temperature experiments (Bureau et al., 2016). In
393 our experiments, the volume of newly formed diamonds is small relative to that of the
394 carbonates, then, since the reaction proceeds by solution and dissolution of carbon in the
395 carbonatitic melt, the isotopic composition of the carbonatite would vary by only a small
396 amount and the new diamond growth would acquire the composition of the carbonatitic melt.

397 Above 1600 °C , which is not relevant to the growth of natural lithospheric diamonds, graphite
398 dissolves in the fluid and provides carbon for diamond growth. In this case the isotopic
399 composition is different (Bureau et al., 2018).

400 If reaction (1) applies, because oxygen is produced, another reaction must occur to react with
401 the newly available oxygen. Graphite would act as an oxygen sink, and possibly as a redox
402 buffer:



404 If reaction (3) does not occur, neither does reaction (1), which can explain why diamonds
405 dissolved when graphite was not included as a starting material in our experiments.

406 This is a general observation, and we here refer to the recent review of Luth et al. (2022) on
407 diamond growth studies performed using various growth media. In all reported experiments
408 on diamond nucleation and growth in silicate melts using graphite as the unique carbon

409 source, neither nucleation nor growth has been reported below 1500 °C. In particular, in the
410 experiments of Fagan and Luth (2011), growth was not observed at 1300 or 1500 °C, but did
411 occur above 1500 °C. Furthermore, except for the study of Bataleva et al. (2012), all
412 experiments reporting diamond nucleation and growth in carbonate-silicate systems used
413 graphite as a starting material (Luth et al., 2022). In the study of Bataleva et al. (2012), when
414 no graphite was added to the carbonate and silicate starting materials, growth on seeds
415 occurred at 1550 and 1650 °C in Pt capsules, whereas dissolution was observed at lower
416 temperatures; in contrast, diamond growth occurred at 1350–1450 °C in experiments using Pt-
417 graphite capsules. All reported experiments that grew diamond in multi-component carbonate-
418 silicate media and/or diamond-forming rocks at temperatures relevant to the lithosphere used
419 graphite either as a starting material or a container (capsule). When graphite was not used in
420 either manner, diamond growth was not observed except when iron was present (Litasov et
421 al., 2014), growth occurred by reduction of carbonates from natural carbonate-bearing
422 eclogites concomitant with Fe²⁺ oxidation of garnets (Kiseeva et al., 2013), or under an
423 electric field (diamond growth on a Pt cathode; Palyanov et al., 2021). Another potential
424 diamond source that is relevant to the Earth and has been investigated experimentally is the
425 carbide system; in the review of Luth et al. (2022), all experiments reporting diamond
426 nucleation and growth involved the presence of graphite and/or metal (Fe, Fe₃C) in the
427 starting materials.

428 This general observation is corroborated by the examination of dissolution experiments;
429 graphite was not added to any of the numerous dissolution experiments reviewed by Luth et
430 al. (2022).

431 It has been proposed that diamond growth and dissolution might alternate within the same
432 system, depending on carbon saturation and redox conditions (Khokhryakov and Pal'yanov,
433 2007). In both the growth and dissolution experiments of this present study, the fluids were
434 carbon-saturated, as indicated by the carbon-rich compositions of the matrix products (Table
435 3). Oxygen fugacity is certainly an important parameter. Palyanov et al. (2013) performed
436 experiments devoted to diamond growth in subduction environments during mantle-slab
437 interactions down to 240 km depth (i.e. at 6 and 7.5 GPa and 1200–1650 °C) in the
438 Mg_{0.9}Ca_{0.1}CO₃-Fe⁰ and Mg_{0.9}Ca_{0.1}CO₃-Fe₃C systems. Within a single carbonate-bearing
439 experimental charge, they observed that diamonds formed both in oxidized Ca-rich carbonate
440 melts and in reduced Fe-C melts, but no dissolution was reported.

441 The above laboratory experiments reproduce the natural growth or dissolution of diamonds in
442 the lithospheric mantle. Indeed, many natural diamonds evidence alternating dissolution and
443 growth events and/or successive growth events in the mantle, as shown by CL mapping of
444 polished diamond sections and/or isotopic profiles within individual diamonds (e.g., Boyd et
445 al., 1994; Zedgenizov et al., 2014; Smart et al., 2016; Gress et al., 2021). If natural diamond
446 growth involves reaction (1), consistent with our present experiments, it requires the presence
447 of components capable of reacting with the O₂ produced by the reaction. Graphite is a good
448 candidate because it is present as inclusions in many diamonds (e.g., Stachel et al., 2022) and
449 possibly as single crystals (e.g., Glinnemann et al., 2003).

450 Other oxygen sinks could be metallic phases and/or sulfides: sulfide inclusions (e.g.,
451 pyrrhotites) are found in many diamonds (see Stachel et al., 2022), and metallic inclusions are
452 present in natural lithospheric diamonds (e.g., Stachel et al., 1988; Bulanova et al., 1998;
453 Daver et al., 2022). The presence of metallic Fe and Fe₃O₄ inclusions was also inferred from
454 X-Ray absorption spectroscopy and Mössbauer spectroscopy analyses of fibrous diamonds
455 (Shiryaev et al., 2010), confirming that diamond formation can occur over a large range of *f*O₂
456 conditions in the lithosphere.

457 The morphologies of the initial diamond seeds and cubes (Fig. 1) recovered from our
458 dissolution experiments recall those produced in diamond dissolution experiments on water-
459 bearing carbonate melts at 5.7 GPa and 1300 °C during 30–75 hours (Khokhryakov and
460 Palyanov, 2015). The dissolution textures observed in our experiments (Fig. 5) may be due to
461 the presence of silicates, excess H₂O (>10 wt.%), and high H₂O/CO₂ ratios (1.6–7.6, Table
462 2), where the highest ratios were associated with the appearance of cuboctahedral shapes for
463 starting diamond cubes (Figure 5I); and showing rounded shapes with trigonal pits (Fig. S6)
464 and preserving (111) faces for starting diamond seeds (Fig. S7). Cuboctahedron shape may be
465 considered as the equivalent stage of morphological dissolution for a initially cubic diamond
466 than dodecahedron is for an initially octahedral diamond. The round final shape of the large
467 initial diamond cube in experiment #38 (Fig 5C) occurred at H₂O/CO₂ = 3.2, but over a
468 relatively long duration of 27 hours. The differences observed between dissolution on seeds
469 and dissolution on cubes also relate to the fact that the surfaces of the starting seeds were
470 oxidized, whereas the surfaces of the broken cubes were not (Fig. 5B–F). As expected,
471 temperature also exerted an important control on resorption, because total lamination was
472 observed at 1400 and 1450 °C, but not at 1350 °C.

473 In their very detailed study, Fedortchouk (2019) combined morphological observations of
474 natural diamonds with descriptions of dissolution and etching experiments. This paper
475 demonstrated the importance of the experimental approach to understanding diamond
476 histories by differentiating resorption that occurred within the mantle source from that which
477 occurred within the kimberlite magma during its ascent to the surface. It is shown that the
478 shape and size of etch pits on diamond surfaces depend on temperature and the H₂O/CO₂ ratio
479 in the fluid, whereas pressure affects the efficiency of diamond crystal-shape transformation
480 from octahedral to rounded, resorbed forms (Fedortchouk, 2019). When the effect of
481 temperature is significant, we observed a change of form at constant pressure (7 GPa), the
482 most significant being the appearance of cuboctahedral shapes in experiment #49 at 1400 °C,
483 also at the highest H₂O/CO₂ ratio (Fig. 5I). The dominant H₂O effect is also shown Fig S7
484 with the preservation of the (111) faces of the starting seeds (see Fedortchouk, 2019).
485 Compared to the classification scheme for natural diamond resorption morphologies proposed
486 by Fedortchouk (2019), our results confirm the destructive character of partial H₂O + CO₂-
487 bearing melts. Following their classification, the dissolution morphologies produced in our
488 experiments fit well with mantle resorption and are applicable to natural diamonds.

489 Our experiments suggest that in nature, a single diamond may record successive growth and
490 dissolution events in similar fluids but under different conditions, in agreement with
491 observations of natural diamonds. These processes may alternate over very short periods of
492 time, as exemplified by our experiments spanning a few hours to a few days. Therefore,
493 protracted episodes of metasomatism between the successive stages are not required to
494 explain observational data from natural diamonds; diamond growth/dissolution in the
495 lithosphere can be rapid. This chimes with recent work predicting the exact same alternation
496 between dissolution and growth during single metasomatic events (Mikhail et al., 2021;
497 Rinaldi et al., 2023). This demonstrates that large portions of the histories of natural diamonds
498 may have been lost during successive, possibly brief, dissolution events.

499

500 ***4.2 Inclusions trapped in diamonds***

501 In our present and previous synthesis experiments, silicates did or did not crystallize
502 depending on the *P-T* conditions, and therefore, when present, syngenetic crystals were
503 trapped as inclusions along with melt (i.e. glass) and/or fluid, or supercritical fluid if the
504 system was beyond total miscibility (Bureau et al., 2016). The high growth rate of synthesized

505 diamonds is the most probable reason for the abundance of entrapped hydrous carbonate-
506 silicate melts and H₂O-CO₂ fluids that represent the diamond-forming media (Bureau et al.,
507 2012, 2016, 2018; Bataleva et al., 2016). It is experimentally shown that all types of diamonds
508 (monocrystalline, fibrous, polycrystalline) can grow in the same media (Bureau et al., 2012,
509 2016). The presence of very thin hydrous silicate films between mineral inclusions and their
510 gem-quality host diamonds (peridotitic or eclogitic) also suggests that the diamond forming
511 medium is composed of water-rich fluids (Nimis et al., 2016). Furthermore, Jablon and Navon
512 (2016) described micro-inclusions trapped along the twinning plane in natural twinned
513 diamonds; the major element compositions of most of these inclusions were similar to those
514 of carbonate-bearing HDFs in fibrous diamonds. They, too, concluded that the mechanism of
515 diamond formation is the same for most diamonds. Therefore, growth kinetics may be a
516 important parameter determining whether a diamond will be monocrystalline or fibrous, with
517 a potential dependence on the presence of water. In natural monocrystalline diamonds,
518 entrapment of the source fluid/melt may not be possible because of the slow growth rates.

519 In our previous study, syngenetic crystals of different compositions were observed as
520 inclusions (phengite, rutile, coesite, carbonates, and diamonds; Bureau et al., 2016), but in our
521 present experiments, no silicate crystals were trapped as inclusions or found within the solid
522 matrix in equilibrium with diamond. Instead, we observed quenched glasses of various
523 compositions between lherzolite and carbonate endmembers (Fig. 4, Table 4). This is likely
524 due to the fast quench rates that surely differ from those experienced by natural diamonds.
525 Whether crystalline or not, these inclusions are syngenetic and represent the diamond
526 crystallization environment.

527 Along with the syngenetic inclusions, we observed pure platinum particles included in a rim
528 in the diamond that corresponds to the initial surface of the diamond seed (this is particularly
529 evident in Fig. 4B, S2). Such inclusions are also observed along with inclusions of fluids,
530 melts, and crystals in a few diamonds grown in previous experiments (Bureau et al., 2012,
531 2016). These particles were from the platinum capsule walls, which could be seen as
532 analogous to the “surrounding rock”. Although platinum might provide a diamond nucleation
533 site in long-term experiments on the kimberlite-graphite system (Pal’yanov et al., 2015), we
534 did not observe any diamonds crystallized on the capsule walls, and therefore we suggest that
535 platinum particles were passively trapped in the diamonds as protogenetic inclusions.
536 Although one might argue that they were previously dissolved in the fluid and would
537 therefore represent syngenetic inclusions, this is very unlikely because the solubility of

538 platinum in silicate melts is on the order of a few tens of ppm (e.g., Blaine et al., 2005;
539 Bezmen et al., 2006).

540 In most cases, it is shown that inclusions trapped at different locations within a single
541 diamond record either one growth event or successive events separated by long periods of
542 time (e.g., Klein-BenDavid et al., 2004; Bulanova et al., 2014). For example, dating of
543 individual growth zones in diamonds from Orapa, Botswana, revealed three distinct ages
544 spanning 3 Ga to 0.3 Ga from core to rim (Gress et al., 2021). In such cases, it is generally
545 assumed that inclusions trapped in the different zones are contemporaneous with growth (e.g.,
546 Koornneef et al., 2017; Gress et al., 2018, 2021), although they might also be protogenetic
547 (e.g., Thomassot et al., 2009; Nestola et al., 2017, 2019). It has been proposed that, because
548 inclusions are small and diffusive processes are fast at mantle temperatures, pre-existing host-
549 rock minerals trapped as inclusions in diamonds chemically and isotopically equilibrate
550 during fluid-mediated growth events, and are therefore “synchronous” but not necessarily
551 syngenetic (Nestola et al., 2017, Stachel et al., 2022). Thus, the lack of syngeneity does not
552 necessarily preclude inclusion-based diamond dating (Smit et al., 2022). Although believed to
553 be exceptional (Stachel et al., 2022), disequilibrium observed among non-touching inclusions
554 (e.g., Griffin et al., 1988; Bulanova, 1995; Wang, 1998) reflects their incorporation either
555 during growth in chemically evolving environments or in different generations (syngenetic
556 and protogenetic) without achieving chemical and isotopic equilibrium. As a last comment,
557 our data agree with Rinaldi et al. (2023) suggestion based on mineral inclusion compositions
558 trapped in natural diamonds that the paragenetic groups used to classify diamonds should not
559 be considered a genetic classification.

560

561 **Conclusions**

562 We experimentally grew and dissolved diamonds compositionally similar to natural
563 lithospheric diamonds; in growth experiments, entrapped inclusions were compositionally
564 similar to those found in natural lithospheric diamonds, although they were not crystalline.
565 The experiments presented here, along with previous ones confirm that all diamonds
566 (monocrystalline, fibrous, polycrystalline, and those formed during high-pressure
567 metamorphism) grow by similar reactions in hydrous carbonate-silicate-rich melts in the
568 mantle. This suggests that their differences are due to different kinetic growth regimes. In our
569 experiments, diamonds underwent dissolution in the absence of graphite, indicating that

570 natural diamonds can dissolve in the mantle within the same fluids from which they grow, and
571 alternating growth and dissolution episodes do not necessarily require protracted periods. This
572 implies that both diamond growth and dissolution involve redox processes governed by
573 reactions that do not conserve oxygen. Growth involves oxygen production and depends on
574 the ability of the system to recombine this oxygen; diamond growth requires that oxygen
575 sinks be present along with the parental fluids/melts. Such sinks could be graphite or metallic
576 and/or sulphide phases, which are commonly observed as inclusions in natural lithospheric
577 diamonds. Alternating growth and dissolution episodes may also be governed by local redox
578 conditions in the mantle. These processes may not be limited to the lithosphere and may also
579 apply to the sublithospheric mantle.

580 Diamond growth and dissolution experiments demonstrate that the histories of diamonds in
581 the mantle are governed by complex processes, and that studying natural diamonds requires
582 careful attention and multiple approaches. Particularly, evidence of the occurrence of multiple
583 dissolution events within a single diamond shows that a significant portion of diamonds'
584 histories may be missing, complicating their reconstruction.

585 Based on our experimental results, we demonstrate that, depending on the growth rate
586 (possibly linked to the amount of water present), diamonds may trap both syngenetic
587 inclusions representing their source medium and protogenetic inclusions that may be totally
588 independent of and much older than the growth medium. In other words, diamonds grow and
589 encapsulate all that surrounds them. No distinction is obvious between these two kinds of
590 inclusions. Therefore, the use of trapped inclusions to date their host diamonds requires
591 careful multi-analytical and structural characterization to prove that they are chemically and
592 isotopically equilibrated, and thus synchronous.

593

594

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610

611 **Appendix A. Supplementary Material**

612 Within this supplementary appendix, the reader can find two tables and height figures.

613 Table S1 describes the details of the experiments, table S2 summarises raw semi-quantitative
614 EDX analysis of matrix glasses from SEM for growth experiments.

615 Fig S1 shows a larger view in sample #52 of the glassy piece showing nano-diamonds
616 growing in the hydrous melt quenched as a mingled glass. Fig S2 shows SEM images of the
617 seed milled with FIB which exhibits a rim of inclusions trapped at the interface between the
618 initial surface of the seed and the grown area, sample #53. Fig S3 shows SEM image of
619 diamond growth on seed of sample #53. Fig S4 shows for sample #55 a SEM image of
620 diamond growth on a broken cube made of one type Ia diamond broken anvil. Fig S5 shows
621 for sample #52 a SEM image of spontaneous nucleation and growth of a diamond (1670°C, 6
622 hours) with octahedral shape. Fig S6 shows for sample #35, a SEM image of the dissolution
623 process at 1450°C 7G GPa, during 5 hours. Fig S7 shows for sample #38 a SEM image of
624 dissolution of a large initial diamond cube (type Ia broken diamond anvil) after 27 hours at
625 1300-1400°C, 7 GPa. Fig S8 exhibits major oxide variation diagrams of inclusions trapped in
626 diamonds compositions (in wt.%, normalized on a volatile-free basis).

627

628

629 **Data Availability**

630

631 Data are included as supplementary materials, they can also be obtained in the Zenodo open
632 data repository (CERN)

633 <https://doi.org/10.5281/zenodo.10361702>

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635

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848 **Figure Captions**

849 **Figure 1**

850 **A.** Optical photograph of diamond pieces from broken diamond anvils; scale is 1 mm. **B.**
851 SEM image of one diamond seed oxidized in air at 1000 °C for 10 minutes; scale is 10 μm. **C.**
852 Open capsule of sample #53 during loading, showing the peridotite sphere in carbonate
853 powder (seeds, graphite, and water are below); scale is 1 mm. **D.** Open capsule of sample #53
854 after the run. The fluid was lost upon opening, the round, yellow central part is mainly
855 composed of seeds, the dark green elongated area is a peridotite (LHERZ) glassy layer, and
856 the surrounding white powder is mostly carbonate. The scale bar is 1 mm.

857 **Figure 2**

858 SEM images showing diamond growth at 7 GPa. Seeds are covered with tiny carbonate
859 crystals agglomerated in clusters. **A.** Sample #51 (1430 °C, 6 hours), showing growth on
860 seeds. **B.** Sample #52 (1670 °C, 6 hours), showing massive growth on the seeds and new
861 diamond crystals attached to seed growths. **C.** Sample #52, showing an octahedral diamond
862 that spontaneously nucleated and grew. **D.** Sample #53 (1400 °C, 24 hours), showing growth
863 on seeds. **E.** Sample #54 (1400 °C, 24 hours), showing an octahedral diamond that
864 spontaneously nucleated and grew. **F.** Sample #55 (1400 °C, 6 hours), showing growth on
865 seeds.

866 **Figure 3**

867 SEM images of sample #52. **A.** A large glassy phase is highly and heterogeneously vesiculated
868 and surrounded by white carbonates. This vermicular texture suggests that a supercritical fluid
869 developed immiscibility during quenching. **B.** Enlarged view of the lower area of this glassy
870 phase, exhibiting small octahedral diamond crystals (“nano-diamonds”) in the process of
871 formation just after nucleation in the single supercritical phase. A larger diamond with growth
872 textures is visible just below this area, and small, white carbonate aggregates are above.

873 **Figure 4**

874 SEM images of diamonds that trapped inclusions during experimental growth (top) before and
875 (middle, bottom) after FIB milling. **A–C.** Sample #53-1. Exposed inclusions are aligned on
876 the initial surface of the seed, marked by the white dashed line in **B.** Different kinds of
877 inclusions are exposed: mixed glasses with vesicles, and bulk white platinum inclusions (Pt,
878 red arrows in **B**). Red crosses correspond to chemical analyses in Table 4. **D–F.** Sample #52-
879 9. Inclusions are melt (now glass) or fluid (now empty voids). **G–I.** Sample #52-5. A large
880 glassy inclusion terminates with an empty void that was initially filled with aqueous fluid. **J–**
881 **L.** Sample #53-6. Massive growth occurred on the top of the seed diamond, and inclusions are
882 aligned along the initial surface of the seed.

883 **Figure 5**

884 SEM images of samples from dissolution experiments. **A.** Sample #32, 1450 °C, 6 hours.
885 Seeds are transformed and exhibit laminations. **B.** Sample #35, 1450 °C, 5 hours. Seeds
886 exhibit advanced lamination. **C.** Sample #35. A large cube of broken diamond that was
887 dissolved and rounded, and no longer shows a clear trigonal shape. **D.** Sample #38, 1300–
888 1400 °C, 27 hours. Seeds are completely foliated. **E.** Sample #38. The surface of a large

889 initial diamond cube is etched and partly dissolved, forming stepped crystal surfaces. **F.**
890 Sample #38. A large, resorbed diamond cube with a rounded final shape that exhibits trigonal
891 and square pits. **G.** Sample #45, 1350 °C, 6 hours. Seeds are partly dissolved. **H.** Sample #44,
892 1350 °C, 6 hours. Seeds are partly dissolved and show pits. **I.** Sample #49, 1400 °C, 6 hours.
893 A partly dissolved cube now showing a cuboctahedral shape, typical of crystal rounding via
894 mantle resorption.

895 **Figure 6**

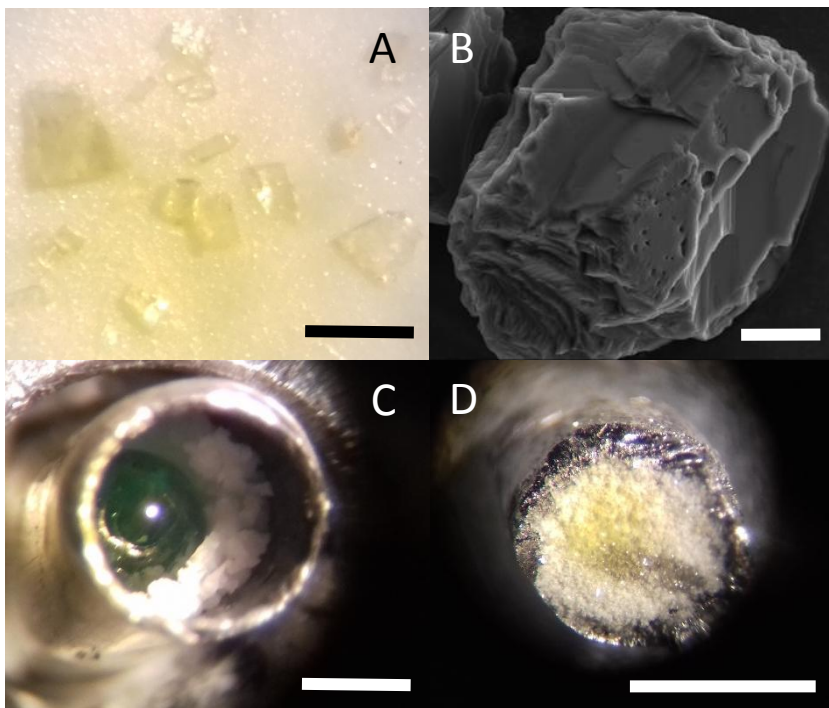
896 The compositions of solid diamond inclusions (blue stars) and matrix glasses (red spots) from
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898 natural fibrous diamonds (grey shaded area; after Weiss et al., 2015 and references therein).

899

900 **Figures**

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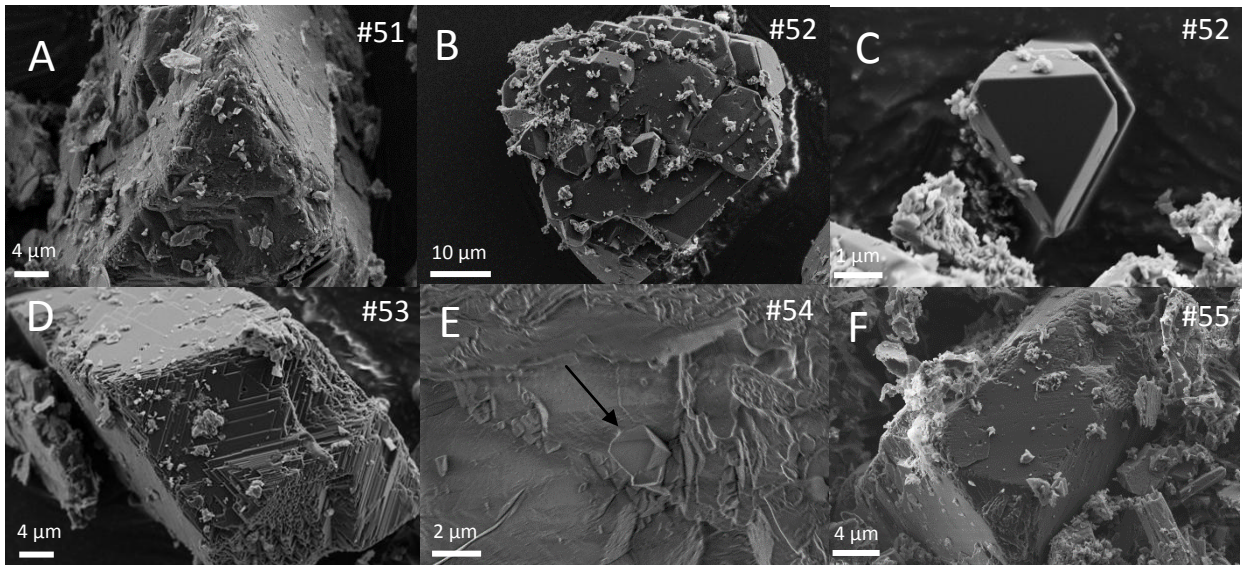


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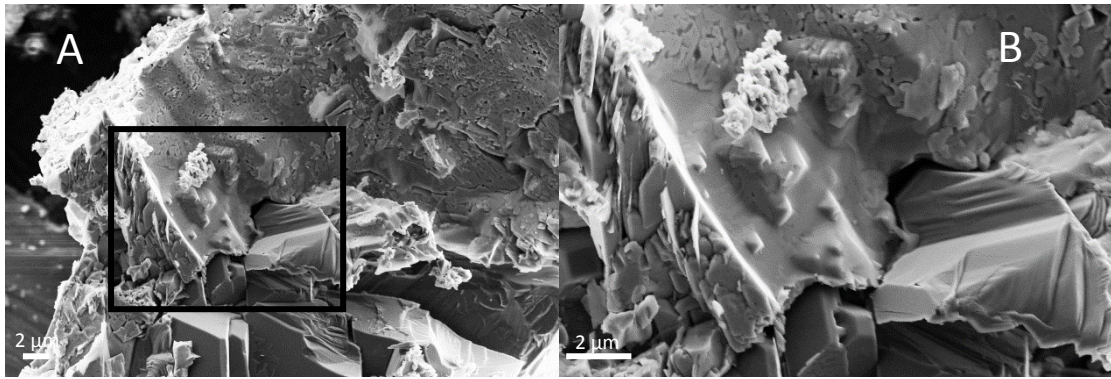
918 seeds.

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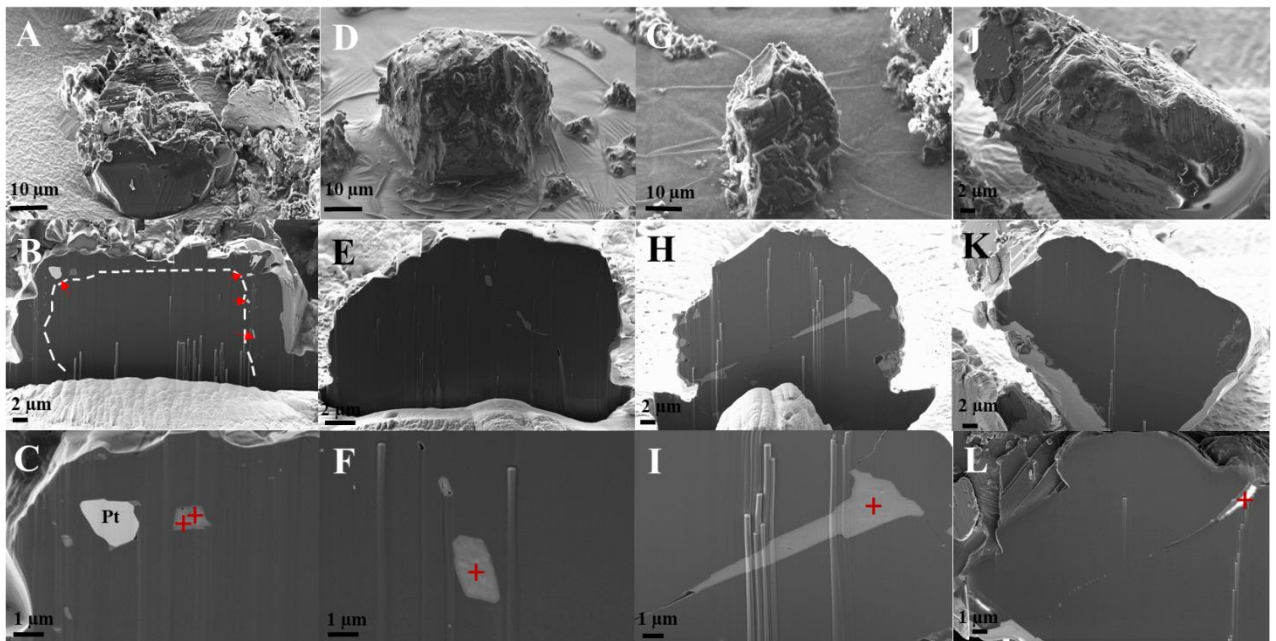
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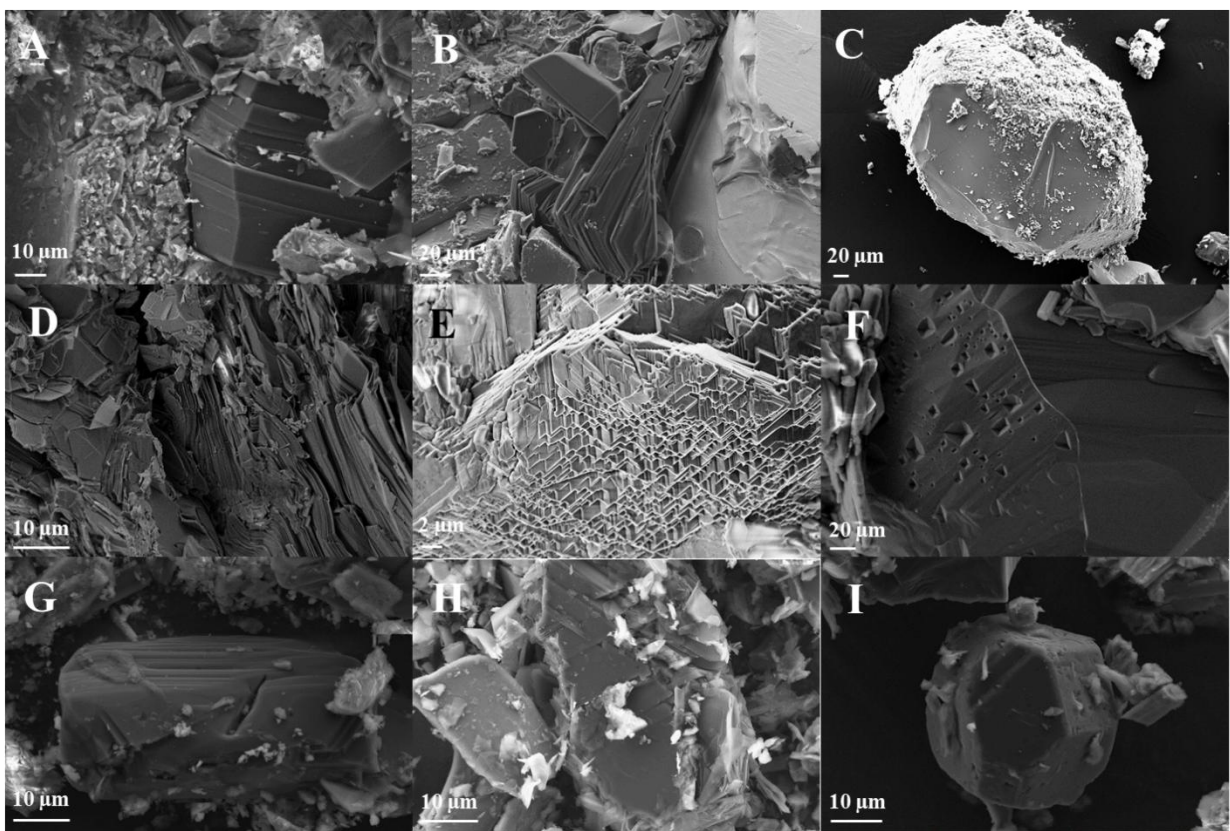


938 aligned along the initial surface of the seed.

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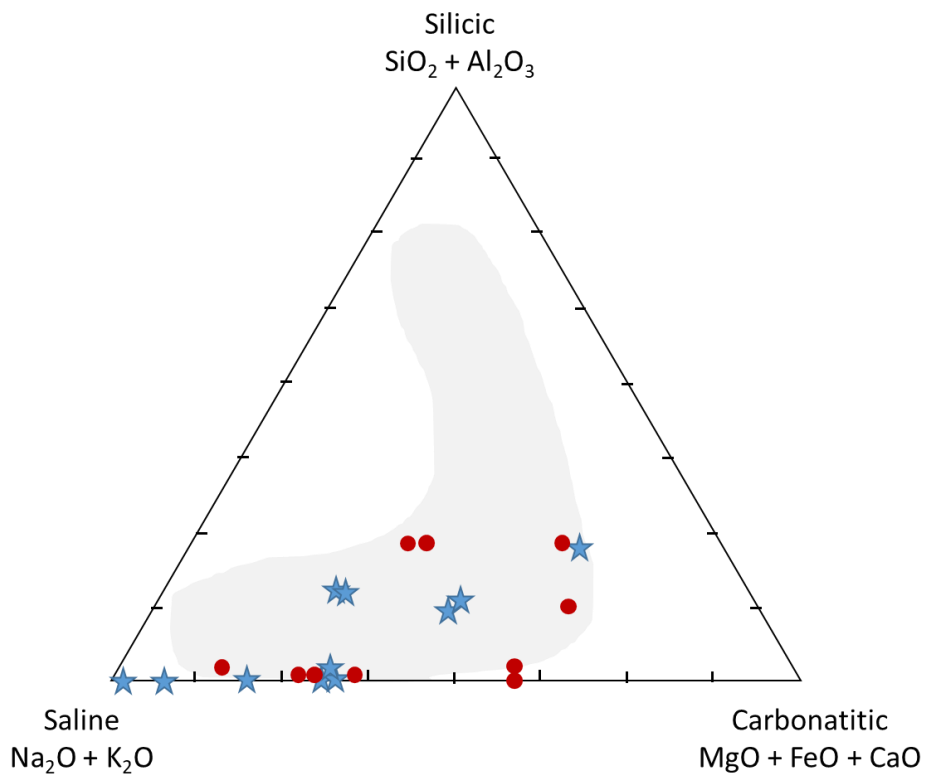
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953 **Figure 6**

954 The compositions of solid diamond inclusions (blue stars) and matrix glasses (red spots) from
955 our experiments plot within the global compositional range of high-density fluids trapped in
956 natural fibrous diamonds (grey shaded area; after Weiss et al., 2015 and references therein).



957

958

959

960 **Table 1:** starting materials

Name	Description	Composition
MORB	Powder of Natural mid-ocean-ridge-basalt, Indian ridge*	DRX analysis: 38.26% $\text{CaAl}_2\text{Si}_2\text{O}_8$ 13.25% $(\text{Mg}_{1.41}\text{Fe}_{0.59})\text{SiO}_4$ 44.72% $(\text{Ca}_{0.8}\text{Mg}_{1.2})\text{Si}_2\text{O}_6$
R_LHERZ	Powder of Natural lherzolite FONB93 from French Pyrénées**	49% olivine ($\text{Fo}_{89.9}$), 33% orthopyroxene ($\text{En}_{89}\text{Fs}_{10}\text{Wo}_1$), 15% Al-rich clinopyroxene ($\text{En}_{4.3}\text{Fs}_{47.4}\text{Wo}_{48.3}$) 3% Al-rich spinel ($\text{Cr}/\text{Cr}+\text{Al}=0.092$) In wt.%: SiO_2 43.26; Al_2O_3 3.23; Fe_2O_3 8.32; MnO 0.12; MgO 37.90; CaO 3.18; Na_2O 0.27; K_2O 0.002; TiO_2 0.12
G_LHERZ	Glass of natural lherzolite	In wt.%.: SiO_2 42.7; Al_2O_3 2.29; Fe_2O_3 8.26; MgO 39.3; CaO 4.4; Na_2O 0.5
Graphite	Powder	C
Ca carbonate	Powder	CaCO_3
Na carbonate	Powder	Na_2CO_3
K carbonate	Powder	K_2CO_3
Seeds	Powder of commercial diamond seeds, 40-60 μm	C

961 *Bureau et al., 2016

962 ** Lorand et al., 2008

963

964

965 **Table 2a:** Summary of the experiments performed at 7 GPa and results to grow diamonds

Exp.	Starting materials	T°C	Time h	Starting H ₂ O/CO ₂	Results (process and products)
Growth					
#51	H ₂ O, Dox, R-LHERZ, K ₂ CO ₃	1430	6	1.8	Aqueous fluid, glass, Carbonates, G, D growth on seeds
#52	H ₂ O, Dox, C, R-LHERZ, G, CaCO ₃	1670	6	2.8	Aqueous fluid, glass, Carbonates, seeds, G, spontaneous D growth, growth on seeds
#53	H ₂ O, Dox, G, C, G-LHERZ, CaCO ₃ , Na ₂ CO ₃ , K ₂ CO ₃	1400	24	4.8	Aqueous fluid, glass, Carbonates, seeds, G, D growth on seeds
#54	H ₂ O, G, C, G-LHERZ, CaCO ₃ , Na ₂ CO ₃ , K ₂ CO ₃	1400	24	4.1	Aqueous fluid, glass, Carbonates, G, D growth on broken cubes, spontaneous D growth
#55	H ₂ O, Dox, G, C, G-LHERZ, Na ₂ CO ₃	1400	6	4.2	Aqueous fluid, glass, Carbonates, seeds, G, D growth on seeds
#50	H ₂ O, Dox, C, G, R-LHERZ, Na ₂ CO ₃	1673	6	4.2	Water leak, no fluid, glass, Carbonates, seeds, G, no D growth
#210*	H ₂ O-NaCl, Dox, G MORB + Natural CaCO ₃	1350	6		Aqueous fluid, glass, G, Carbonates. Slight D growth on seeds and spontaneous D growth

966 Notations: Exp. Experiment; Dox oxidized diamond seeds; G graphite; MORB Mid Ocean
 967 Ridge Basalt; R-LHERZ powder of lherzolite; G-LHERZ glass of lherzolite; C diamond
 968 cubic pieces (100-200µm). * from Bureau et al., 2016. (H₂O/CO₂) ratio by weight, graphite
 969 not included. In the case of runs #52 and #50 performed at HT, the ratio may reach 0.8 and
 970 0.5 assuming all G is dissolved in the fluid. As G is observed in the quenched products, the
 971 ratio is higher.

972

973 **Table 2b:** Summary of the experiments performed at 7 GPa and results to dissolve diamonds

Exp.	Starting materials	T°C	Time h	Starting H ₂ O/CO ₂	Results (process and products)
Dissolution					
#31	H ₂ O, Dox, R-LHERZ, CaCO ₃ , Na ₂ CO ₃ , K ₂ CO ₃	1450	6	4.3	Aqueous fluid, Carbonates, glass, resorbed seeds
#32	H ₂ O, Dox, R-LHERZ, CaCO ₃ , Na ₂ CO ₃ , K ₂ CO ₃	1450	6	2.2	Aqueous fluid, Carbonates, glass, resorbed seeds
#35	H ₂ O, Dox, C, MORB, CaCO ₃ , Na ₂ CO ₃ , K ₂ CO ₃	1450	5	1.6	Aqueous fluid, Carbonates, glass, resorbed seeds Resorption of the D cubes
#38	H ₂ O, Dox, C, MORB, CaCO ₃ , Na ₂ CO ₃ , K ₂ CO ₃	1300-1400	27	3.2	Aqueous fluid, Carbonates, glass, resorbed seeds Resorption of the D cubes
#44	H ₂ O, Dox, R-LHERZ, K ₂ CO ₃	1350	6	4.1	Aqueous fluid, Carbonates, glass, resorbed seeds
#45	H ₂ O, Dox, R-LHERZ, CaCO ₃	1350	6	3.8	Aqueous fluid, Carbonates, glass, resorbed seeds
#49	H ₂ O, Dox, C, R-LHERZ, Na ₂ CO ₃	1400	6	7.6	Aqueous fluid, Carbonates, glass, resorbed seeds

974 Notations: Exp. Experiment; Dox oxidized diamond seeds; G graphite; MORB Mid Ocean
 975 Ridge Basalt; R-LHERZ powder of lherzolite; G-LHERZ glass of lherzolite; C diamond
 976 cubic pieces (100-200µm). * from Bureau et al., 2016. (H₂O/CO₂) ratio by weight, graphite
 977 not included. In the case of runs #52 and #50 performed at HT, the ratio may reach 0.8 and
 978 0.5 assuming all G is dissolved in the fluid. As G is observed in the quenched products, the
 979 ratio is higher.

980

981

982 **Table 3a:** Some semi-quantitative SEM-EDX representative analysis of the matrix glasses
983 associated to the seeds for samples #54, #53 and #55 (diamond growth). A strong
984 heterogeneity of carbon is observed, this is due to the strong contamination due to the
985 diamond crystals present close to or below the small pieces of glasses. Raw compositions are
986 presented in Table S2, they show that the total is ranging from 64 to 100. Water is omitted
987 because water cannot be analysed with SEM and because of the strong uncertainties of CO₂
988 contents, a calculation of water contents by difference to 100 is not possible. Compositions
989 are calculated to obtain a total of 100 wt%. The strong enrichment in Na for sample #55
990 reflects the fact that only NaCO₃ was added for this experiment. Same remark for sample #52
991 and CaCO₃. For samples #54, #53, the 3 carbonates were present. For sample #52, one
992 analysis is performed in a glassy area exhibiting nano-diamonds and vermiculates that witness
993 a strong water degassing during the quench (Figure 3), the four last analyses are performed in
994 the same glassy area.

Grow th	#54	#54	#53	#53	#53	#53	#55	#55	#52	#52	#52	#52	#52
	GoS	GoS	GoS	C G	GoS	GoS	GoS	G	GoS	GnD	vG	vG	vG
SiO ₂	26.6	23.5	12.3	0.45	36.3	23.5	20.8	7.18	8.98	12.0	13.9	18.9	4.95
Al ₂ O ₃	8	2	5	0.00	0	7	9	0.86	0.00	3	1	2	0.55
	7.60	1.10	2.79	0.00	2.49	3.84	1.75	18.5	0.00	0.00	0.00	0.00	0.40
Na ₂ O	5.06	3.46	0.74	7.93	1.60	2.64	3.91	5	0.00	0.00	0.00	0.00	0.00
	19.4	16.4			16.3	13.6	11.6	10.8		12.9	13.1	14.5	16.2
MgO	3	3	9.32	3.21	4	8	0	3	4.51	9	4	3	3
	12.6		15.1		12.7					0.00	0.00	0.00	0.00
K ₂ O	3	2.47	5.84	3	2.37	1	0.00	0.00	0.00	0.00	0.00	0.00	0.00
									31.8	22.5	20.3	22.1	16.2
CaO	0.71	5.67	0.00	6.68	8.94	1.09	0.25	1.66	2	3	0	9	3
	27.8	47.3	68.9	66.6	31.9	42.4	61.5	60.9	54.7	51.1	51.5	42.6	66.7
CO ₂	9	4	5	0	6	7	9	2	0	4	7	6	7
	100.	100.	100.	100.	100.	100.	100.	100.	100.	100.	100.	100.	100.
Total	00	00	00	00	00	00	00	00	00	00	00	00	00

995

996

997 Table 3b: Some semi-quantitative SEM-EDX representative analysis of the matrix glasses
 998 associated to the seeds for samples and #35, #38 (diamond dissolution). Same legend as for
 999 Table 3a

Dissolut ion	#35	#35	#35	#38	#38	#38	#38	#38	#38	#38	#38	#38	#38
	G	G	G	G	GoS	G	G	G	G	G	G	G	G
SiO₂	21.2 9	21.0 7	0.00	19.9 9	2.96	31.0 2	12.1 1	38.9 4	2.56	24.7 0	27.0 5	10.1 9	21.2 9
Al₂O₃	4.09	3.73	0.00	5.65	1.02	8.91	4.77	11.7 6	1.31	7.85	8.11	4.02	4.09
Na₂O	4.42	3.89	12.8 3	1.99	0.55	2.39	0.67	2.62	1.72	2.27	1.45	1.52	4.42
MgO	18.9 4	18.7 7	16.4 1	4.65	8.99	6.61	8.19	8.85	23.1 0	11.3 5	6.93	9.07	18.9 4
K₂O	3.32	4.96	6.26	0.00	0.00	0.66	4.31	0.78	0.86	1.51	0.00	1.77	3.32
CaO	13.3 0	19.0 9	29.6 8	8.41	13.2 9	14.7 9	2.82	12.6 3	35.2 3	11.9 9	11.0 5	22.7 0	13.3 0
CO₂	34.2 4	27.9 8	34.8 2	59.3 1	73.2 0	34.6 2	66.7 0	23.5 4	35.2 2	39.8 4	44.5 7	50.7 3	34.2 4
Total	100. 00	100. 00	100. 00	100. 00	100. 00	100. 00	100. 00	100. 00	100. 00	100. 00	100. 00	100. 00	100. 00

1000 Notations: G Glass; GoS Glass on Seeds; C G carbonate glass; GnD glass with nano-
 1001 diamonds; vG vermiculated glass

1002 **Table 4:** Volatile-free semi-quantitative analysis of the inclusions trapped in diamond seeds.
 1003 For these analyses, due to the strong carbon contamination from diamond, carbon is not taken
 1004 into account. Positions of the analysis are indicated on figure 4 with red crosses. No minerals
 1005 are trapped, all inclusions are glassy, for #53 (1 and 2) the inclusion is highly vesiculated.

	#53-1 1	#53-1 2	#53-6 3	#52-5 4	#52-9 5
SiO₂	37.60	23.20	47.35	19.82	30.74
Al₂O₃	6.81	3.74	9.28	0.00	0.00
Na₂O	0.00	0.00	0.00	0.00	0.00
MgO	35.44	44.08	20.98	9.94	12.89
K₂O	13.28	14.74	22.39	0.00	1.59
CaO	6.87	14.23	0.00	70.24	54.79
Total	100.00	100.00	100.00	100.00	100.00

1006

1007

