1 **Revision 1**

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3 **Positively-oriented trigons – a unique resorption feature of diamonds from Snap**

4 **Lake kimberlite dyke, Canada**

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13 **Abstract**

14 The role of fluid(s) in the formation of different lithological facies of kimberlites is still

15 poorly understood. The ambiguity of composition of kimberlite melts hampers

16 understanding the composition of volatiles, their depth of exsolution and the effect on

17 magma ascent and fragmentation. Recent estimates of H_2O and CO_2 solubility in

18 kimberlite-like magmas suggest very shallow exsolution of fluid, while many features of

19 kimberlites indicate the presence of significant fluid fraction at depth. Deep magmatic

20 fluid produces negative trigonal etch pits on natural diamonds, the characteristics of

21 which depend on the temperature and composition of the fluid. Positively-oriented

22 trigonal etch pits are very rare on natural diamonds, and are likely a feature of resorption

23 events unique to only some kimberlite magmas. Here we present the first systematic

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24   study of positively oriented trigonal etch pits on natural diamonds from Snap Lake

25   kimberlite dyke, Northwest Territories, Canada. The study used 91 micro-diamonds

26   selected from a population of 251 diamonds representative of all six kimberlite litho-

27   facies identified in the Snap Lake dyke. We established that unlike the majority of

28   diamonds from kimberlite pipes in the Northwest Territories, every studied Snap Lake

29   diamond shows positively-oriented trigons. These trigons cover the whole diamond

30   surface starting from the {111} faces and continuing over the resorbed edges. They

31   overprint negatively-oriented trigons and modify them into hexagons. Atomic force

32   microscopy obtained detailed geometry of 154 positive trigons on fourteen diamonds.

33   Three distinct trigon morphologies dependent on the type of the crystal lattice defect

34   were recognized. The point-bottomed shape and positive correlation between the depth

35   and diameter of the individual pits suggest a high CO_2 content in the fluid. Comparison

36   with the existing experimental data on positive trigons implies resorption at low-pressure

37   conditions in the 800-1000°C temperature range by trapped magmatic fluid after the dyke

38   emplacement. The intensity of this late resorption event (and the size of the positive

39   trigons) increases from the dyke contact with the country rock into the interior of the

40   dyke. Such a late resorption event is absent in the majority of kimberlites, which form

41   pipes, and might be a specific feature of hypabyssal kimberlite bodies (dykes). The

42   absence of positive trigons on diamonds from the majority of kimberlites suggests very

43   quick magma cooling below ~800°C after the pipe emplacement, precluding the

44   development of any late resorption features. Our study shows that for kimberlitic magmas,

45   for which mineral chemistry is unable to provide a robust record of magmatic fluid,

46   morphological details of dissolution features on the surface of diamond and other mantle-

47 derived minerals can serve as a fluid proxy. Better constrains of the pressure, temperature

48 and oxygen fugacity of the reversal in the trigon orientation on diamond may help to

49 reconstruct the emplacement path of geologically diverse kimberlite bodies.

50 Keywords: diamond resorption, kimberlitic fluid, Snap Lake kimberlite, trigonal etch pits,

51 atomic force microscope

52

53 **Introduction**

54 Diamonds recovered from kimberlites and other deep-seated mantle magmas show a

55 combination of growth and resorption features on their surfaces. Resorption features are

56 products of diamond dissolution both in the mantle source and in the host magma, while

57 the morphology of these features depends on the conditions in both natural diamond

58 environments. Experiments show that kimberlitic fluid plays a major role in the

59 development of resorption features on diamonds (e.g. Fedortchouk et al. 2007). Thus,

60 better understanding the controls of diamond resorption morphology provides a tool for

61 examining the formation and composition of magmatic fluid during the ascent and

62 emplacement of kimberlite magma, independent of the uncertainty of the primary

63 composition of kimberlite melt (e.g. Moussallam et al. 2016).

64 The common resorption features found on the surfaces of natural diamonds

65 include: etch pits of trigonal, tetragonal, and hexagonal shape, terraces, laminae, hillocks

66 of rounded or sharp shape, and shallow depressions, circular pits, and corrosion

67 sculptures (e.g. Robinson 1979). Trigonal etch pits of negative orientation are a common

68 resorption features (Fig. 1a), which develop via fast propagation of step 2 (Fig. 1a)

69 consisting of carbon atoms doubly bonded to the crystal lattice. Positive trigons are very

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70 rarely reported on natural diamonds (Robinson 1979) and develop via fast propagation of

71 step 1 consisting of carbon atoms trebly bonded to the crystal lattice (Fig. 1a) (Yamaoka

72 et al. 1980). Negative trigons are routinely observed in experimental studies conducted at

73 pressures of 1 GPa and higher (Arima and Kozai 2008; Fedortchouk 2007; Khokhryakov

74 and Palyanov 2010) and occure on diamonds from every kimberlite locality and any

75 kimberlite facies. Their maximum size depends on the temperature of the reacting fluid

76 (e.g. Fedortchouk 2015), while shape and abundance is very different in H_2O and CO_2

77 fluids (Khokhryakov and Palyanov 2010, Fedortchouk 2007). Zhang (2016) further

78 examined how the CO_2 content of the reacting fluid affects the relative lengths of the

79 positive (L_{pos}) and negative (L_{neg}) walls of trigonal etch pits (Fig. 1b) where C-O-H fluid

80 with <40 mol% CO_2 produces negative trigons with flat bottomed (f/b) and regular shape;

81 fluid with 40-90 mol% CO_2 produces f/b negative trigons with truncated corners; and

82 fluid with >90 mol% CO_2 produces pointed-bottomed (p/b) trigons and transitional

83 trigon-hexagon form. In addition, dissolution in CO_2-rich fluid produces negative trigons

84 with apparent positive correlation between the diameter and depth of the negative trigons,

85 while in H_2O-rich fluid this relationship is absent (Fedortchouk 2015).

86 Positive trigons have been very rarely reported on natural diamonds (Robinson,

87 1979). Typically, they occur on rare diamond crystals with extremely etched surfaces

88 found in parcels where other diamonds do not show positive trigons. In addition to the

89 positive trigons, these diamonds also show other very rare features, such as transverse

90 hillocks (of the opposite orientation compared to the common hillocks and imbricate

91 wedge markings (Fig. 1c,d). Positive trigons are the most common feature observed in

92 experiments conducted at 0.1 MPa but have been also produced at high pressures (high-P)

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93   (e.g. Yamaoka et al., 1980, Harris and Vance, 1974). The causes of the reversal in the

94   orientation of trigonal etch pits on diamonds are still not well understood. Experimental

95   studies observed the change from negative to positive trigons at high oxygen fugacity

96   (fO_2) at 0.1 MPa (Evans and Sauter 1961) and at 1.5 GPa (Yamaoka et al. 1980) and

97   temperatures of 900°C and 1130°C respectively. These authors proposed that T and fO_2

98   control the orientation of the trigons via stabilization of doubly bonded atoms of step 2

99   (Fig. 1a) with oxygen complexes (Evans and Sauter 1961; Yamaoka et al. 1980).

100   Similarly, T and fO_2 control a reversal in the orientation of tetragonal etch pits on {100}

101   diamond faces (Fedortchouk and Canil 2009). However, experiments at 5-7 GPa and

102   1400 – 1750°C (Khokhryakov and Palyanov 2010) produced positive trigons in dry

103   carbonate melt and reversed to negative trigons after addition of 8 wt% of H_2O at the

104   same fO_2, which allowed the authors to propose that the CO_2:H_2O ratio is the main

105   control for the trigon orientation. On the contrary, Harris and Vance (1974) recorded at

106   0.1 GPa and 1050°C negative trigons in carbonate-rich composition and positive in more

107   H_2O rich compositions. Thus, the existing experimental data does not give an explicit

108   answer on the causes of change in trigon orientation. A theoretical model by Angus and

109   Dyble (1975) proposes that formation of positive trigons requires preferential removal of

110   triply bonded carbon atoms, which requires more energy than removal of the doubly

111   bonded atoms that accompanies negative trigon formation. Better understanding of the

112   controls of the trigonal etch pit orientation on diamonds may help to constrain

113   crystallization conditions of kimberlite magma.

114   Snap Lake kimberlite dyke (Northwest Territories, Canada) is a unique example

115   of a kimberlite body, where all diamonds show positive trigons. What makes Snap Lake

116   so unique? Are the positive trigons on Snap Lake diamonds a product of deep (high-P)

117   resorption due to the different kimberlite emplacement conditions than the majority of

118   kimberlites, or do they represent a different resorption event happening at shallow near-

119   surface conditions in a hypabyssal kimberlite body as opposed to the typical kimberlite

120   that bursts through the earth's surface to form a pipe? Can we deduce the crystallization

121   conditions of the Snap Lake kimberlite(s) using the morphology of positive trigons? We

122   address these questions via a detailed study of the positive trigons on diamonds from

123   different litho-facies of the Snap Lake kimberlite dyke. Employment of an Atomic Force

124   Microscope (AFM) for study of diamond surface features allowed us not only to detect

125   the presence of small-sized trigons, but also to examine how the shape and dimensions of

126   the trigons vary throughout the kimberlite body. Here we report the first systematic study

127   of the morphology and characteristics of positive trigons on natural diamonds recovered

128   from known sections of a single kimberlite body. The study aims to understand what

129   factors control the presence, morphology, and dimensions of the positive trigons, and to

130   examine their distribution within the Snap Lake dyke in order to better understand the

131   role of volatiles in kimberlite emplacement.

Background geology and diamond samples

133   The Snap Lake Mine is located in the southeastern part of the Slave Craton about

134   220 km northeast of Yellowknife, Canada (Fig. 2a) (Fulop et al. 2017). The age of the

135   kimberlite, 535-523 Ma, was determined by Rb-Sr dating of phlogopite (Agashev et al.

136   2001; Heaman et al. 2003, 2004). The country rocks comprise granodiorites, tonalities

137   and granites of the Defeat Pluton Suite (2610 – 2590 Ma) and metavolcanic rocks of the

138   Yellowknife Supergroup (Stubley 2000). There is no evidence of a thermal aureole in the

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139   country rocks adjacent to the intrusion (Gernon et al. 2012). The orebody of the Snap

140   Lake kimberlite is a segmented hypabyssal dyke dipping 12-15º towards the northeast

141   flanked by a series of subparallel sheets (Gernon et al. 2012; Kopylova et al. 2010). The

142   thickness of the dyke ranges from 0.1 to 15 m, but typically ~ 3-5 m (Gernon et al. 2012).

143   The Snap Lake dyke is filled with a coherent hypabyssal kimberlite HK

144   (following the terminology of Scott Smith 2013) with minor kimberlite breccia facies

145   dominated by granite or amphibolite xenoliths (Kopylova et al. 2010; Mogg et al. 2003).

146   Two models have been proposed to explain the compositional variation and the

147   emplacement of the Snap Lake dyke. A two magma batches model suggests that the two

148   different litho-facies (the main olivine-rich and phlogopite-poor facies, and the

149   subordinate phlogopite-abundant facies) evolved from two magma batches which co-

150   mingled and intruded almost simultaneously (Gernon et al. 2012; Ogilvie-Harris et al.

151   2008; Field et al. 2009). A single magma batch model suggests formation of one

152   hypabyssal kimberlite with different degrees and forms of deuteric alteration and crustal

153   contamination responsible for the generation of six sublitho-facies from HK1 to HK6,

154   which is supported by petrographic, geochemical, and mineral chemistry data (Mogg et al.

155   2003, Kopylova et al. 2010, Fulop et al. 2017).The middle of the dyke is composed of the

156   freshest phlogopite-poor HK1 and HK2 facies, with more altered HK5 surrounding

157   granitoid xenoliths. The progressively more altered phlogopite-rich HK3, HK4 and HK6

158   facies developed at the contact with the country-rock granitoids (Fig. 2b).

159   Our study used 251 micro-diamonds provided by De Beers Canada Inc with the

160   maximum diamond size 1.7 mm. We selected 91 diamonds that are ≥300 μm in size and

161   can be handled under an optical microscope (diamonds <300 μm are mostly fragments

162   and provide no information about resorption of crystal faces) and that are not fragments

163   or show at least 50% of the crystal faces, so that crystal morphology can be studied. Our

164   study focused on octahedral or tetrahexahedral (THH) diamonds, since cubic diamonds

165   are lacking experimental data on their resorption. The selected 91 diamonds comprise 78

166   octahedral and 13 THH diamonds, which represent all litho-facies at Snap Lake: HK1 (25

167   diamonds), HK2 (25 diamonds), HK3 (4 diamonds), HK4 (14 diamonds), and HK5-HK6

168   (23 diamonds). AFM was conducted on 16 diamonds representing all litho-facies and

169   Fourier transform infrared spectroscopy (FTIR) measurements were obtained for 82

170   diamonds (including few fragments). The diamonds were recovered at Saskatchewan

171   Research Council (SRC) by caustic fusion at 500°C in $Na(OH)_2$. Use of test synthetic

172   diamonds with every batch confirms no etching during the recovery process.

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Analytical methods

Optical and Scanning Electron Microscopy (SEM)

The diamonds were initially studied using a stereomicroscope to examine the general morphology, color, color intensity, and the presence of inclusions. Further examination of growth/resorption features on diamond surfaces and grouping into morphological types used a stereomicroscope and reflected light on a petrographic microscope with partially closed aperture stop for increased contrast. We classified diamonds with more than 10% of {111} faces preserved as an octahedron, and with less than 10% of {111} faces preserved − as a tetrahexahedron (THH). The morphologies of twinned diamonds and aggregates were classified based on the dominant shape of the constituent crystals. The octahedral diamonds were further divided into several morphological groups based on: (1) trigonal vs. ditrigonal shape of {111} faces, and (2) the type of resorption features on the {111} face including step faces, etch pits (negative and positive trigons, trigon-hexagons, hexagons), and laminae (shield-shaped, serrate- or irregular-shaped). SEM examination of 29 representative diamond crystals further refined the morphological grouping. Positive trigons larger than 1 μm were recorded with an optical microscope. The presence of positive trigonal etch pits smaller than 1 μm on the studied diamonds was confirmed with SEM. Diamonds selected for SEM were cleaned with a boiled HNO_3-H_2SO_4 mixture (3:5 volume proportion) at ~200°C for at least 30 min followed by ultrasonic cleaning in distilled water for 15 min and were then coated with gold. We used a Field Emission Scanning Electron Microscope (FE-SEM) Hitachi S-4700 FEG at the Institute of Research in Materials, Dalhousie University, with accelerating voltage of 10-15 kV and beam current of 15 μA.

Atomic Force Microscopy (AFM)

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196   The AFM study of 16 diamonds (positive trigons were studied on 14 diamonds) was

197   conducted using a Veeco Multimode 8 atomic force microscope at the Department of Earth

198   Sciences, Dalhousie University. The diamonds were cleaned with aqua regia (HNO_3-HCl in 1:3

199   volume proportion) to remove the gold coating and other possible metal impurities, and then

200   with a HNO_3-H_2SO_4 mixture (3:5 volume proportion) to remove the carbon from the SEM tapes

201   and other dirt. During both treatments, the diamond crystals were boiled at ~200°C for at least 30

202   minutes. Finally, the crystals were cleaned in distilled water in an ultrasonic bath for 15 minutes.

203   Prior to the AFM measurements, all selected diamonds were photographed at 100× and 200×

204   (500× for minute features such as positive trigons) magnification using a petrographic

205   microscope. Each diamond was mounted on a steel disk with an adhesive pad, in an attempt to

206   orient the examined {111} face horizontally. In the case of uneven or fragmented crystals glue

207   from the adhesive tape was placed under the crystal to minimize the tilt. All AFM images were

208   collected using a J scanner (Bruker, maximum coverage 125 μm × 125 μm × 5 μm in x, y and z

209   dimensions), operated in contact ScanAsyst mode using ScanAsyst Air Probes (Bruker, silicon

210   tip on nitride lever, spring constants 0.4 to 0.8 N m^{-1}, resonant frequency 50-90 kHz, nominal tip

211   radius of curvature 2 nm). The scanned areas ranged from 5 × 5 μm to 80 × 80 μm. The scanning

212   frequency was dependent on the size of the scanned areas: we used 0.2 – 0.4 Hz for areas greater

213   than 30 × 30 μm and 0.5 – 0.8 Hz for smaller areas to optimize the match between the trace and

214   retrace lines. All the scans were conducted with 512 sample lines.

215   The images were analyzed using NanoScope software (v8.10) after a "plane fit" function

216   to remove the tilt in both x and y directions. The parameters of individual etch pits (diameter,

217   depth, shape, wall angles) were examined on cross-section profiles using the section analyses

218   tool. A profile was drawn from the middle of a trigon side to the opposite vertex. For each

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219   diamond we routinely measured 8-20 positive trigons. We measured the diameter of the pits as a

220   distance between the middle of one side and the opposite vertex, the depth of the pits as the

221   distance between the {111} face and the bottom of the pit, and the wall angles ($\alpha_{\{111\}}$) as the

222   angle between the plane of the wall micro-face and (111) plane. Each parameter comprises an

223   average of the three measurements for the profile drawn from each of the three pit vertexes. The

224   pits were classified based on the bottom and wall morphology (Fig. 3) (see also Zhang et al.

225   2015). The three sources of uncertainties in AFM measurements include: (1) the AFM

226   uncertainty (1.4% in x, y and z directions); (2) a possible tilt of the {111} diamond face; (3) the

227   subjective selection of the measurement (the placement of the profile line and the markers used

228   for the measurements). We estimate the total uncertainty <2.5% for diameter measurement, <6%

229   for depth measurement, and <4º angle measurement (Table 1) (see also Zhang et al. 2015; Li

230   2017).

231   **Fourier transform infrared spectroscopy (FTIR)**

232   Nitrogen content and aggregation state were obtained with Fourier transform infrared

233   spectroscopy (FTIR) for the 14 diamond crystals with positive trigons studied by AFM (Table 2).

234   Diamond crystals were mounted on the edge of a glass slide with the (111) faces perpendicular to

235   the FTIR beam. We used Thermo-Nicolet Nexus 470 FTIR spectrometer fitted with a

236   Continuμm infrared microscope equipped with a KBr beam splitter at the Department of Earth

237   and Atmospheric Sciences at the University of Alberta. Absorption spectra were measured in

238   transmission mode in the range of 4000–650 cm^{-1} with a resolution of 8 cm^{-1}, and were collected

239   for 200 s through a 100 ×100 μm^2 aperture. Sample spectra were first baselined using the Basic

240   Macro of the OMNIC 32 software suite. A normalized Type II diamond spectrum used as a

241   standard for background correction was subtracted from the spectra to convert absorbance to

242 absorption coefficient. After the conversion, sample spectra were de-convoluted into the A, B

243 and D components using least square techniques (for more details see Stachel et al. 2006, Boyd

244 et al. 1994, 1995). The detection limit for nitrogen is about 10 ppm and the analytical precision is

245 10-20 % of the concentration (Stachel et al., 2006).

246 **Diamond etching experiments**

247 The temperature limit for the development of any resorption features on diamond at 0.1 MPa was

248 tested in experiments conducted at 700°C and 800°C at oxygen fugacity (fO_2) corresponding to

249 that of the air (logfO_2 = -0.68) using a box furnace and at fO_2 corresponding to that of CO_2 gas

250 (logfO_2 = -2.85 at 700°C and -2.79 at 800°C) using a vertical-tube gas-mixing furnace in the

251 Department of Earth Sciences at Dalhousie University. In both cases, natural octahedral

252 diamonds (weight ~0.5 – 1 mg) with minimal natural resorption were imaged with AFM to

253 record any pre-existing features, placed inside Al_2O_3 crucible, and covered with a synthetic

254 mixture of 45 mol% Na_2CO_3 and 55 mol% NaCl corresponding to the eutectic composition with

255 melting temperature 632°C. The run duration was 2.5 hours in the air runs and 3 hours in CO_2

256 runs. After the experiments the diamonds were recovered, cleaned following the described above

257 procedure, and imaged with optical microscope and AFM to detect any new features on the

258 diamond's surfaces.

259 **Results**

260 **General morphologies and negative trigons of Snap Lake diamonds**

261 We divided the octahedral and THH diamonds from Snap Lake into twelve

262 morphological groups based on the shape of {111} faces (trigonal vs. ditrigonal) and the type of

263 the surface resorption features. The details of morphological groupings and full description of the

264 negative trigonal etch pits will be reported elsewhere. Here we focus on the features of the

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265   positive trigons. The 14 diamonds for which positive trigons were studied with AFM are all

266   colourless octahedra, which represent eight morphological groups illustrating both mantle-

267   derived and kimberlite-induced resorption (Fig. 4). The number of negative and positive trigons

268   analyzed on each diamond is reported in Table 2. AFM study of the negative trigons revealed

269   that they all have regular flat-bottomed (f/b) shape, diameter < 45 μm, and depth < 1 μm. The

270   unique feature of all negative trigons on Snap Lake diamonds is their truncated corners to a

271   degree that makes them transitional into hexagons (Fig. 5e,f). Many negative trigons contain

272   small positive trigons inside (Fig. 5b,c,f) or along their perimeter (Fig. 5a,d).

273   **Positive trigons on Snap Lake diamonds**

274   All 91 studied diamonds from Snap Lake kimberlite have positive trigons, which cover

275   both octahedral and THH faces. While negative trigons are limited to {111} faces, positive

276   trigons occur also on the rounded THH edges of resorbed diamonds (Fig. 6a,b). Positive trigons

277   are distributed uniformly (Fig. 6a) or show a patchy uneven distribution of larger and smaller

278   trigons in different areas of {111} faces (Fig. 6c). They also enhance the development of wormy

279   depressions (ruts) (Fig. 6h). The majority of the positive trigons have a diameter <3 μm, which

280   correlates with their depth (Fig. 7). Diamonds from HK1 and HK2 facies have larger trigons and

281   only two of the fourteen diamonds (SNP8-9 and SNP9-1) have positive trigons with diameters

282   larger than 4 μm (up to 13 μm). Less resorbed diamonds have positive trigons with diameter ≤

283   0.5 μm and depth < 20 nm, which could only be detected using SEM or AFM. The diameter of

284   the positive trigons roughly correlates with the kimberlite lithology and decreases from fresh

285   (HK1 and HK3) and slightly altered facies (HK2 and HK4) towards highly altered facies (HK5

286   and HK6) (Fig. 8). Diamonds from HK2 show three larger diameter trigons in Fig. 8, which

287   might be a result of an overprint of pre-existing pits. We found no correlation between the size of

288   the positive trigons and the general diamond morphology.

289   The detailed AFM study of 154 positive trigons on 14 Snap Lake diamonds (reported in

290   Supplementary Table 1) allowed us to divide them into four groups (Fig. 9):

291   1) point-bottomed (p/b) trigons with an irregular outline and walls consisting of multiple

292   micro-steps (Fig. 9b)

293   2) trigons similar to above but the bottoms are curved (c/b) and not pointy; the walls have

294   V or U shape; trigons with simple corners have more rounded bottoms (Fig. 9c), while trigons

295   with multiple corners have flatter bottoms (Fig. 9d)

296   3) asymmetric trigons with straight outlines and regular steps along one of the walls,

297   shallower pits are f/b (Fig. 9e) and deeper pits are p/b (Fig. 9f)

298   4) trigons with truncated corners and transitional trigon-hexagons with straight walls and

299   well-defined f/b shape (Fig. 9g)

300   The shape (f/b vs. p/b) of the small embryonic trigons with diameter <1 μm and depth <

301   100 nm (e.g. Fig. 9a) cannot be reliably distinguished with the scanner used. P/b is the most

302   prevalent shape of the positive trigons on Snap Lake diamonds; only transitional trigon-hexagon

303   pits show regular f/b shape and most likely represent an overprint of positive trigons on small

304   pre-existing negative trigons.

305   There are two distinct trends on diameter vs. depth plots (Fig. 7) where p/b trigons form a

306   steeper slope and c/b trigons form a shallower slope. P/b trigons are the most common and occur

307   on the majority of the studied diamonds. They form the only trigon type on the three diamonds

308   from HK1 and HK2 (SNP2-1, SNP8-9, SNP3-6). Asymmetric trigons are limited to only two

309   diamonds from HK1 and HK2 (SNPO13-2, SNPO9-1), that also show highly intensive etching

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310   of {111} face. These trigons evolve from f/b to p/b as their depth increases. Curved c/b trigons

311   appear alone (SNP3-8) or in combination with p/b trigons (SNP6-1, SNP9-1, SNPO12-13,

312   SNPO12-8). Trigons with flatter bottoms mostly occur on diamonds from phlogopite-rich altered

313   facies HK4 and HK6. Trigon-hexagon f/b pits were only observed on two diamonds from the

314   altered facies at the contact with granitoids of HK5 (phlogopite-poor) and HK6 (phlogopite-rich)

315   kimberlites (SNP5-3, SNPO12-2). The wall type of the studied trigons shows an evolution from

316   U to V to Y as the depth of the trigons increases on the same diamond (Supplementary table 1).

317   **Experimental result**

318   In this study we only focused on the presence of absence of any changes to diamond surface after

319   heating in sodium carbonate-chloride melt at $700^{o}C$ and $800^{o}C$ and $logfO_2$ = -0.68 (air), -2.79

320   and -2.85 (CO_2). Fig. 10 shows that etching at $800^{o}C$ developed positive trigons on diamond

321   surface easily seen on the micro-photographs taken under an optical microscope. After $700^{o}C$

322   runs, no changes were visible under an optical microscope, but AFM showed development of

323   positive trigons only in the run with air. AFM imaging detected no changes on diamond surface

324   after the run at $700^{o}C$ in CO_2.

325   **Discussion**

326   **Where do positive trigons form?**

327   In experiments positive trigons form over a wide pressure range: at 0.1 MPa (Evans and Sauter

328   1961), 0.1 GPa (Harris and Vance, 1974), 1.5 GPa (Yamaoka et al. 1980), 5-7 GPa

329   (Khokhryakov and Palyanov 2010). The distribution patterns of positive trigons on natural

330   diamonds from Snap Lake kimberlite support the conclusion of Robinson (1979) these positive

331   trigons are a feature of the latest resorption event, which can overprint previous resorption.

332   Indeed, positive trigons cover the whole diamond surface including the rounded resorbed faces

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333   (Fig. 6) and bottoms of the larger negative trigons. Furthermore, the AFM images obtained in our

334   study demonstrate that positive trigons on Snap Lake diamonds overprint all the pre-existing

335   negative trigons. They are present on the bottoms, along the sides or in the center of the negative

336   trigons, and add truncated corners to all the negative trigons resulting in their transition to trigon-

337   hexagon and hexagon shapes (Fig. 5a-f). Our results demonstrate that the presence of positive

338   trigons on Snap Lake diamonds records conditions of a particular late resorption event.

339   Reversal in the trigon orientation manifested in the formation of trigon-hexagonal and

340   hexagonal pits has been observed during diamond dissolution in CO_2-rich fluid (Fedortchouk et

341   al. 2007, Khokhryakov and Palyanov, 2010). Zhang (2016) found that truncation of the corners

342   of negative trigons starts when CO_2 constitutes more than 50 mol% of the fluid and hexagons

343   form at CO_2 content >95 mol%, where the ratio $L_{pos}/(L_{pos}+L_{neg})$ can be used to quantify the

344   degree of trigon to hexagon transformation (Zhang, 2016). If the development of positive walls

345   inside the negative trigons on Snap Lake diamonds was controlled by the CO_2 content of the

346   magmatic fluid, as in the Zhang (2016) experiments, both negative and positive walls of

347   truncated trigons and trigon-hexagon pits would have developed simultaneously during a single

348   resorption event so that the $L_{pos}/(L_{pos}+L_{neg})$ ratio would be independent of the diameter of the pit.

349   In such a case, a correlation with the kimberlite lithology would be expected. If the formation of

350   the positive walls overprinted the original shape of a negative trigon, then the $L_{pos}/(L_{pos}+L_{neg})$

351   ratio would show a negative correlation with the trigon diameter. Fig. 11a shows the correlation

352   for the studied Snap Lake diamonds, where the smaller the diameter of the negative trigons the

353   larger is their $L_{pos}/(L_{pos}+L_{neg})$ ratio. Fig. 11a also shows a different trend for HK1 and HK2

354   diamonds with a larger $L_{pos}/(L_{pos}+L_{neg})$ ratio for the same diameter compared to the diamonds

355   from the other lithologies. Diamonds from HK1 and HK2 also have a larger absolute value of

16

356 L_{pos} (Fig. 11b) and larger diameter of the positive trigons (Fig. 8) indicating the higher intensity

357 of the resorption event forming positive trigons in the HK1 and HK2 kimberlite facies. The

358 micro-features on the positive walls of truncated negative trigons on Snap Lake diamonds are

359 different from those produced in CO_2-rich dissolution experiments by Zhang (2016) (Fig. 5g-i).

360 In these experiments, the truncation by positive wall development was accompanied by a

361 curvature of the negative trigon walls (Fig. 5g-i). All this evidence confirms that trigon-

362 hexagonal and hexagonal pits on Snap Lake diamonds are not a product of resorption in more

363 CO_2-rich fluid but a combination of two resorption events – an early (high-P) event, that

364 produced negative trigons typical for the majority of kimberlitic diamonds, and the later (low-P)

365 event, that produced positive trigons and truncated negative trigons, a specific feature of the

366 Snap Lake diamonds.

367

368 **Application to Snap Lake kimberlite system**

369 This late resorption event in the Snap Lake kimberlite most likely occurred after dyke

370 emplacement, while the kimberlite was still at magmatic temperature. Experimental studies of

371 diamond etching help to constrain the temperature of this late resorption event responsible for the

372 development of the positive trigons on Snap Lake diamonds. Yamaoka et al. (1980) estimated

373 1130°C as a reversal temperature from negative to positive orientation of trigonal etch pits at 1.5

374 GPa. Slightly lower reversal temperature between 900 and 1000°C obtained by Evans and Sauter

375 (1961) at more oxidized conditions cannot be explained by the difference in f_{O_2}, which according

376 to Yamaoka et al. (1980) should increase the reversal temperature at higher f_{O_2}. Most likely the

377 reason for this discrepancy is in erroneous f_{O_2} estimates in Yamaoka et al. (1980). Their study

378 used a double capsule technique with an oxide buffer in the outer capsule and diamond crystals

17

379   in the small capsule inside. Such an approach requires reaching equilibrium in the system, which

380   cannot be achieved until the diamonds are completely oxidized. As long as the diamond

381   oxidation reaction proceeds the f_{O2} would be somewhere between the value imposed by the oxide

382   buffer and the value of the CCO buffer. Thus, we suggest 900 - 1000°C as the maximum

383   temperature for the development of positive trigons on Snap Lake diamonds, which is slightly

384   below the ~1100°C crystallization temperature of kimberlite magma (Fedortchouk and Canil

385   2004).

386   The minimum T for this resorption event is the T limit for diamond resorption. According

387   to the experiments of Yamaoka et al. (1980) this is 800°C. A number of studies (e.g. Evans and

388   Sauter 1961, Phaal 1965) observed diamond etching at temperatures as low as 650°C but only in

389   pure oxygen, and above 950°C in CO_2 flow. Our experiments conducted at yet more oxidized

390   conditions than that of natural kimberlites show that at 700°C resorption occurs in the air but

391   does not proceed in pure CO_2 gas corresponding to $log\ fO_2$ = -2.85 (Fig. 10). We propose the

392   temperature of the late resorption event forming positive trigons on diamonds from Snap Lake

393   dyke between 800 – 1000°C.

394   Kimberlite magma typically arrives at the earth's surface with substantial amounts of

395   magmatic fluid formed via CO_2 degassing accompanying assimilation of silicate xenoliths by

396   carbonate-rich magma (e.g. Russell et al. 2012; Stone and Luth 2016) as well as from

397   decompressional degassing of H_2O and CO_2 in the last few km of the ascent (Moussalam et al.

398   2016). Extensive serpentinization and other alteration reactions observed in kimberlites can be a

399   product of reaction with the magmatic fluid (e.g. Mitchell 2013) or can result from a substantial

400   contribution from meteoric fluid (e.g. Stripp et al. 2006). The high content of phlogopite in Snap

401   Lake kimberlite, especially closer to the contact with the country-rock granitoids (HK3,4,6 facies)

18

402   was interpreted as a product of alteration by deuteric fluid (Fulop et al. 2017). The dimensions of

403   the positive trigons on diamonds can help to examine the nature of the fluid during the late

404   resorption event at Snap Lake. The diameter of the trigonal etch pits on diamonds depends on T

405   and the duration of the diamond reaction with fluids (Fedortchouk 2015). The largest diameters

406   of the positive trigons on diamonds from our study are limited to the two freshest facies (HK1

407   and HK2, Fig. 8) present in the interior parts of the intrusion far from the contact with the

408   country rock. Diamonds from the facies closer to the contact with the granitoid (HK3,4,6) or

409   surrounding large granitoid xenoliths (HK5) (Fig. 2b) show very small sizes of the positive

410   trigons. This implies longer interaction with fluid possibly at higher T in the interior parts of the

411   intrusion. In combination with the estimated high T of the late resorption event, this would

412   strongly support magmatic fluid as the resorption agent. We propose that at depth, during the

413   ascent of the Snap Lake kimberlite magma to the surface, magmatic fluid gave rise to resorption

414   features typical for all kimberlite-hosted diamonds (such as negative trigons). After the

415   emplacement, the fluid was trapped in this hypabyssal intrusion close to the surface, and resulted

416   in the later resorption event manifested in the development of the positive trigons on Snap Lake

417   diamonds, which is not present on diamonds from both volcaniclastic and coherent lithologies in

418   kimberlite pipes in Ekati Mine, Canada, and Orapa kimberlite cluster, Botswana (Fedortchouk et

419   al 2010; Fedortchouk et al. 2017).

420   However, Snap Lake may not be that unique. Many diamonds from this study have so

421   small trigons, which were only detected with SEM, and would have been overlooked during a

422   routing examination under an optical microscope even with 50x objective. Indeed, AFM

423   scanning of octahedra micro-diamonds from unknown Yakutian sources used for diamond

424   dissolution experiments have shown presence of positive trigons on some diamonds

19

425   (Fedortchouk, unpublished data). Furthermore, there is only very limited data on diamonds from

426   kimberlite sheets. Some of them are not diamondiferous, others are too thin to be mined.

427   Diamonds from kimberlite pipe are lacking such widespread development of positive trigons,

428   and only show them on rare individual diamonds in association with other very rare features (Fig.

429   1 c,d). It is possible that a late resorption event forming positively-oriented trigons is a feature of

430   kimberlite sheets. The fluid responsible for the late resorption is clearly not related to

431   serpentinization that occurs at low temperatures around 130-400°C (Afanasyev et al. 2014) and

432   creates strongly reducing conditions (McCollom and Seewald 2013).

433   The shape of negative trigons on diamond can be used to deduce the $H_2O:CO_2$ ratio in the

434   reacting fluid (Fedortchouk 2015). Twelve out of fourteen diamonds studied by AFM show two

435   types of positive trigons: 1) p/b trigons with straight walls (which evolve from small flat almost

436   f/b pits, Fig. 9a,b), and 2) curved-bottom trigons (Fig. 9c,d). Both types of trigons form distinct

437   positive trends of diameter vs. depth (Fig. 7), the slope of which is steeper for p/b trigons and

438   shallower for curved/bottomed trigons. Study of the negative trigons from diamond dissolution

439   experiments (Fedortchouk 2015) suggests that such trigon morphologies and the positive

440   correlation between the diameter and the depth are indicative of CO_2-rich fluid. However,

441   applicability of this correlation to positively oriented trigons has yet to be confirmed

442   experimentally. Negative trigons on Snap Lake diamonds show very regular f/b shape of the

443   diamonds that suggests predominantly aqueous fluid in the earlier high-P resorption event during

444   the magma ascent (Fedortchouk et al. 2007; Fedortchouk 2015). The shape of the positive trigons

445   would agree with the fluid evolution towards more CO_2-rich composition after the emplacement

446   during crystallization of the dyke, given that CO_2 effect on the shape of positive and negative

447   trigons is similar. Assimilation reactions between the carbonate component of the melt and

448   silicates from the granitoids may have provided an additional CO_2 input. Alternatively, the

449   difference in the shape of negative and positive trigons can be related to the different dissolution

450   conditions during the earlier (high-P) and late (near-surface) resorption events. Luttge (2006)

451   proposed that at conditions close to equilibrium (small ΔG), crystal dissolution proceeds via

452   formation of flat-bottomed "pancake" pits growing around kinks associated with point defects.

453   When the conditions are far from equilibrium (at higher ΔG), dissolution opens the hollow cores

454   at screw dislocations forming deeper p/b etch pits. Pressure has a large control on the stability of

455   the diamond lattice. A decrease in pressure shifts the system very far from an equilibrium

456   (towards larger ΔG). The presence of large diameter but shallow negative trigons and the

457   absence of such shallow positive trigons on Snap Lake diamonds might be due to much higher

458   ΔG at low P.

459   Development of both f/b and p/b asymmetric trigons is limited to only three diamond

460   crystals, of which two have no other trigon types (Fig. 7, SNPO13-2, SNPO9-1). This is likely

461   not due to the external conditions of the resorption but the internal properties of these diamond

462   crystals and their defects. Finally, truncated and trigon-hexagonal pits were only observed on

463   diamonds from kimberlite facies very close to the contact with large xenoliths (HK5) or country-

464   rock (diamond SNPO12-2 from HK6, Fig. 7). Incomplete transformation of very small (<2 μm in

465   diameter) negative trigons into positive trigons, suggests a very short duration of resorption and

466   confirms the increase in the resorption intensity towards the center of the dyke.

467

Mechanism of diamond resorption and development of positive trigons

469   Review of the existing experimental data shows uncertainty in the conditions required for

470   formation of negative vs. positive trigons on diamonds during dissolution. Yamaoka et al. (1980)

471   following Evans and Phaal (1962) and Phaal (1965) propose that T and fO_2 affect the activity of

472   oxygen in stabilizing certain directions of the diamond lattice via C – O – C bridges (Fig. 1a).

473   However, evolution from negative trigons to hexagons and positive trigons was observed in

474   experiments at the same T, P, and fO_2, but conducted in H_2O-rich and CO_2-rich systems

475   (Fedortchouk et al. 2007; Khokhryakov and Palyanov 2010; Zhang 2016) emphasizing the role

476   of water in the orientation of etch pits on diamond. Angust and Dyble (1975) developed a

477   theoretical model based on an ideal configuration of diamond crystal lattice (Fig. 12), which

478   proposed that development of negatively-oriented trigons is expected for the diamond lattice

479   because this mechanism involves preferential removal of carbon atoms bonded to fewer of the

480   surrounding atoms. On the contrary, the development of positively-oriented trigons requires

481   conditions with no preferential removal of triply-bonded and doubly-bonded carbon atoms. This

482   could be achieved when the diamond crystal lattice becomes very unstable, such as at very low P

483   or very high fO_2. Since diamond is a high-pressure carbon phase, emplacement of kimberlite

484   magma at shallow levels could create conditions where the diamond lattice becomes extremely

485   unstable, the activation energy needed to remove triply-bonded and doubly-bonded atoms

486   become similar, and positive trigons start to form. This agrees well with the observed occurrence

487   of positive trigons on diamonds from the shallow Snap Lake kimberlite sheet but their absence

488   on diamonds from Ekati kimberlite pipes both located in Slave craton of Canada (e.g.

489   Fedortchouk et al. 2010). Indeed, most of the experiments on diamond dissolution or oxidation

490   report positive trigons at 0.1 MPa and negative trigons at elevated P. While low pressure is not

491   the only possible way to destabilize diamond and produce positive trigons, it appears to be the

492   most likely cause in natural kimberlitic systems.

493   Positive trigons developed in high-P experiments accompany features of extreme

494   diamond etching. For example, naturally occurring rare diamonds with positive trigons shown on

495   Fig. 1c,d best resemble figure 6 from Yamaoka et al. (1980) study, which shows experimentally

496   oxidized diamond at 4 GPa at the extreme oxidation conditions corresponding to $Mn_2O_3 - MnO_2$

497   oxygen fugacity buffer. We propose that there are at least two different natural mechanisms that

498   produce positive trigons on diamonds by enhancing removal rate of triply-bonded carbon atoms

499   (Angus and Dyble 1975). First, the extreme oxidation (and / or heating) experienced by

500   diamonds in the mantle source, can explain presence of extremely rare individual diamond

501   crystals found in parcels where other diamonds show no positive trigons. Second, near-surface

502   resorption event can overprint all diamonds with tiny positive trigons similar to the ones

503   described here on Snap Lake diamonds. As SEM imaging becomes a routine step in

504   characterization of diamond parcels, the proposed here association of positive trigons with

505   diamonds from kimberlite sheets can be further tested.

506

507   **Implications**

508 It has been shown that the common resorption features of diamonds such as negative trigons,

509 hexagons, and circular pits show strong association with the CO_2/CO_2+H_2O ratio and

510 temperature of the oxidizing fluid and hence may serve as a proxy of fluid in kimberlite systems

511 (e.g. Fedortchouk et al. 2007; Zhang et al. 2015). By expanding this method to positive trigons in

512 this study, we confirmed that the trigon diameter correlates with T and duration of the resorption

513 event and may be used to examine the thermal history of fluids during crystallization of a

514 kimberlite body. We also confirmed that the correlation of the diameter and the depth established

23

515   for negative p/b trigons exists for positive p/b trigons, and likely results from the CO_2/CO_2+H_2O

516   ratio in the fluid.

517   Our findings place some constraints on the crystallization conditions of different kimberlite

518   bodies. In the Snap Lake dyke we identified two diamond resorption events produced by the

519   kimberlite magma. The early high-P resorption occurred in all kimberlite litho-facies during the

520   ascent and produced negative trigons. The later low-P resorption event produced the positive

521   trigons on all studied Snap Lake diamonds at near-surface conditions (pressures approaching 0.1

522   MPa) by high-temperature magmatic fluid ($\sim 800 – 1100^{o}C$). Since experiments at 0.1 MPa show

523   development of positive trigons as fast as within one hour (Fedortchouk, unpublished data),

524   absence of positive trigons on diamonds from the majority of kimberlite bodies suggests very

525   quick magma cooling below $\sim 800^{o}C$ precluding the development of any late resorption features.

526   The presence of positive trigons on diamonds indicates unusual emplacement conditions. Further

527   studies are needed to clarify if such resorption events are a feature of kimberlite sheets, which do

528   not breach the surface and better preserve magmatic volatiles.

529   Solubility of H_2O and CO_2 in kimberlite-like magma (Moussallam et al, 2016) suggests

530   exsolution of both volatiles only very close to the surface within the last 2-5 km of the ascent.

531   This could drive the explosive eruption and magma fragmentation, but would not explain the

532   extremely fast ascent of kimberlites (e.g. Canil and Fedortchouk 1999, Kelley and Wartho 2000,

533   Sparks et al., 2006). A reaction between silicate xenoliths and carbonate-rich kimberlite magma

534   may exsolve CO_2 starting as deep as 70-80 km ($\sim$2.5 GPa) (Russell et al. 2012, Stone and Luth

535   2016) to drive the fast kimberlite ascent. However, the features of negative trigons on diamonds

536   indicate notable variation in CO_2/CO_2+H_2O ratio of the fluid (Fedortchouk 2015), which in some

537   kimberlites is less than 50 mol% CO_2 (Zhang, 2016). This study observed a change from the f/b

538   shape of the negative trigons (typical for H_2O-rich fluids) to p/b shape of the positive trigons

539   (typical for CO_2 –rich fluids), which may suggest CO_2 increase in the fluid during the ascent

540   through the last kilometers. Indeed, experimentally determined solubilities of H_2O and CO_2 show

541   an increase in CO_2 exsolution closer to the surface (Moussallam et al, 2016). Yet, it is not clear if

542   H_2O component in kimberlitic fluid introduced due to decompressional degassing at 2-5 km

543   below the surface could be responsible for all the observed resorption on kimberlitic diamonds.

544   Our study shows that a better understanding of P, T, and fO_2 of the reversal in trigon orientation

545   may provide a tool to establish the depth of exsolution and changes in the composition of

546   kimberlitic fluid. Since the development of the positive trigons is limited by P and T, this would

547   probably provide the most robust record of crystallization conditions and emplacement

548   mechanism of kimberlite magma.

549   Further testing of the proposed association of positive trigons on diamonds with kimberlite

550   sheets, can have important implications for diamond exploration. It is important to distinguish

551   between kimberlite sheets and hypabyssal kimberlites of the root zone of kimberlite pipes at the

552   early stage of a drilling program. This impact the assessment of the ore reserve calculation and

553   modelling of kimberlite body. Perhaps examining for the presence of positive trigons on micro-

554   diamonds under SEM can help better planning of drilling expenses.

555

556   **Acknowledgments**

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566

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684

685   **Figure Captions**

686

687   Figure 1 - A) Positive and negative trigons on {111} diamond face and their formation due to

688   relative rates of propagation of step 1 (positive trigons) and step 2 (negative trigons) (after

689   Yamaoka et al. 1980). B) Truncated trigon on {111} diamond face with lengths of positive (L_{pos})

690   and negative (L_{neg}) micro-faces shown (after Zhang 2016). C) Natural diamond of unknown

691   geographic origin with abundant positively oriented trigons and imbricate wedges. D) Natural

692   diamond (octahedral macle twin) with positively oriented trigons, fine transverse hillocks on

693   resorbed dodecahedral surfaces and imbricate wedge markings.

694

695   Figure 2 – Geology of the Snap Lake kimberlite dyke. A) The location and shape of the dyke. B)

696   Distribution of the six kimberlite litho-facies within Snap Lake dyke relative to the contact with

697   the two types of the country-rocks (after Fulop et al. 2017).

31

698

699   Figure 3 – Classification approach for trigonal etch pits on diamond based on the bottom and

700   wall geometries (modified from Zhang et al. 2015). The pits are defined as having V type walls

701   when $[\alpha_1 - \alpha_2] < 1^o$ and as f/b when $[\beta_1 - \beta_2] < 1^o$.

702

703   Figure 4 – Microphotographs of sixteen Snap Lake diamonds, on which positive and negative

704   trigons were studied by AFM.

705

706   Figure 5 - Development of positively-oriented micro-faces on negative trigons on Snap Lake

707   diamonds (A – F) and in experiments at 1250^oC and 1 GPa with CO_2-rich fluids (modified from

708   Zhang 2016) (G – I). A – C – development of truncated corners of trigons on Snap Lake

709   diamonds; D – F – trigon-hexagons and hexagons on Snap Lake diamonds; G – I – negative

710   trigons from the Zhang (2016) experiments showing curvature of trigon edges accompanying

711   truncation of the corners with positive micro-faces.   The degree of corner truncation

712   ($L_{pos}/L_{pos}+L_{neg}$ ratio) increases from (G) to (I). All images are AFM scans.

713

714   Figure 6 – Distribution of positive trigons on Snap Lake diamonds: on resorbed THH faces (a,b),

715   on {111} faces (c, d, e, f), and inside the negative trigons (g, h). The boxes on the left images

716   show the location of the close-up area displayed on the right. a, b, c, e, g – microphotographs; d,

717   f, h – AFM images.

718

719   Figure 7 – Diameter vs. depth of positive trigons on Snap Lake diamonds from different

720   kimberlite litho-facies based on the AFM measurements. For the trigons types see Fig. 9.

721

722   Figure 8 – Diameter of positive trigons on diamonds from different litho-facies of Snap Lake

723   kimberlite. A) The choice of a representative maximum trigon size of the positive trigons on

724   individual diamonds studied with optical microscope or SEM. B) Use of the above approach to

725   establish the proportions of diamonds with a maximum size of the positive trigons $\leq$ 3 μm,

726   between 3-5 μm, and $\geq$ 5 μm in each kimberlite facies. C) The diameter range of the individual

727   positive trigons measured with AFM on Snap Lake diamonds from different kimberlite facies.

728

729   Figure 9 – Morphological types of positive trigons on Snap Lake diamonds based on the AFM

730   profiles.

731

732   Figure 10 – Etching of diamond surface in experiments with Na_2CO_3-NaCl melt conducted at 0.1

733   MPa, 700°C and 800°C and fO_2 corresponding to that of the air and pure CO_2 gas. AFM images

734   of diamond surface before and after the runs at 700°C show development of positive trigons in

735   air and but no sign of resorption in CO_2.

736

737   Figure 11 - A) The relationship between the ratio $L_{pos}/L_{pos}+L_{neg}$ and the diameter of trigonal etch

738   pits on diamonds from different kimberlite facies showing that the trend defined by diamonds

739   from the fresh HK1-HK2 kimberlites is different from the trend defined by diamonds from more

740   altered kimberlites (HK4, 5, 6). B) L_{pos} values of etch pits measured on individual diamonds

741   from different kimberlite facies (the diamond labels are shown along the x-axes, the number of

742   measured pits is shown in brackets).

33

743

744   Figure 12 - Atom removal process and development of trigonal etch pits on {111} diamond

745   surfaces (from Angus and Dyble 1975) showing development of a negative trigon (B) from an

746   arbitrary hole in the outer layer (A) after sequential removal of doubly-bonded atoms. X is a top

747   layer atom bonded to the layer below. 0 is a top layer atom not bonded to the layer below.

748

749

750   **Tables**

751   Table 1 – Summary of the sources of uncertainty in AFM measurements.

752   Table 2 –AFM details and nitrogen data for the fourteen diamond from Snap Lake kimberlite

753   studied by AFM.

754

755   **Electronic supplementary materials**

756   ES table 1 – AFM data of Snap Lake diamonds

757

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760

761

Table 1 Summary of the sources of uncertainty in AFM measurements.

Parameter	Source of uncertainty		Total uncertainty
	Systematic Errors	Random Errors	
Diameter	AFM uncertainty after calibration (1.4%); Tilt of the raw image before Plane Fit operation (<0.2%)	Subjective selection of measurements (0.6%)	<2.5%
Depth	AFM uncertainty after calibration (1.4%); the remaining tilt and the compression of the image after the Plane Fit operation (<4%);	Subjective selection of measurements (0.3%)	<6%
$\alpha_{\{111\}}$	AFM uncertainty after calibration (<2º); Tilt of the raw image and the remaining tilt after the Plane Fit operation (<1º)	Subjective selection of measurements (<1º)	<4º

Table 2 AFM details and nitrogen data (from FTIR) for the fourteen diamond from Snap Lake kimberlite studied on AFM.

Diamond	Lithology	# sides scanned	# of etch pits scanned			Nitrogen defects	
			positive trigons	negative trigons	hexagons	N total ppm	%B defects
SNP2-1	HK1	1	11	3	1	526.9	20.3
SNP8-9		1	11	0	0	1027.3	16.5
SNPO13-2		1	9	2	1	893.2	36.0
SNP3-6	HK2	1	8	0	1	46.6	97.9
SNP3-8		1	10	2	0	1431.9	4.8
SNP6-1		1	19	8	0	321.6	1.1
SNP9-1		2	11	0	1	344.2	54.2
SNPO9-1		1	8	0	0	388.3	58.2
SNP1-9	HK4	1	10	0	0	1021.9	50.4
SNP10-9		1	9	1	2	502.5	26.7
SNP5-3	HK5	1	12	11	2	100.4	57.6
SNPO12-2	HK6	1	9	4	0	178.5	37.5
SNPO12-3		2	18	2	0	246.6	43.1
SNPO12-8		1	9	1	0	17	65.9
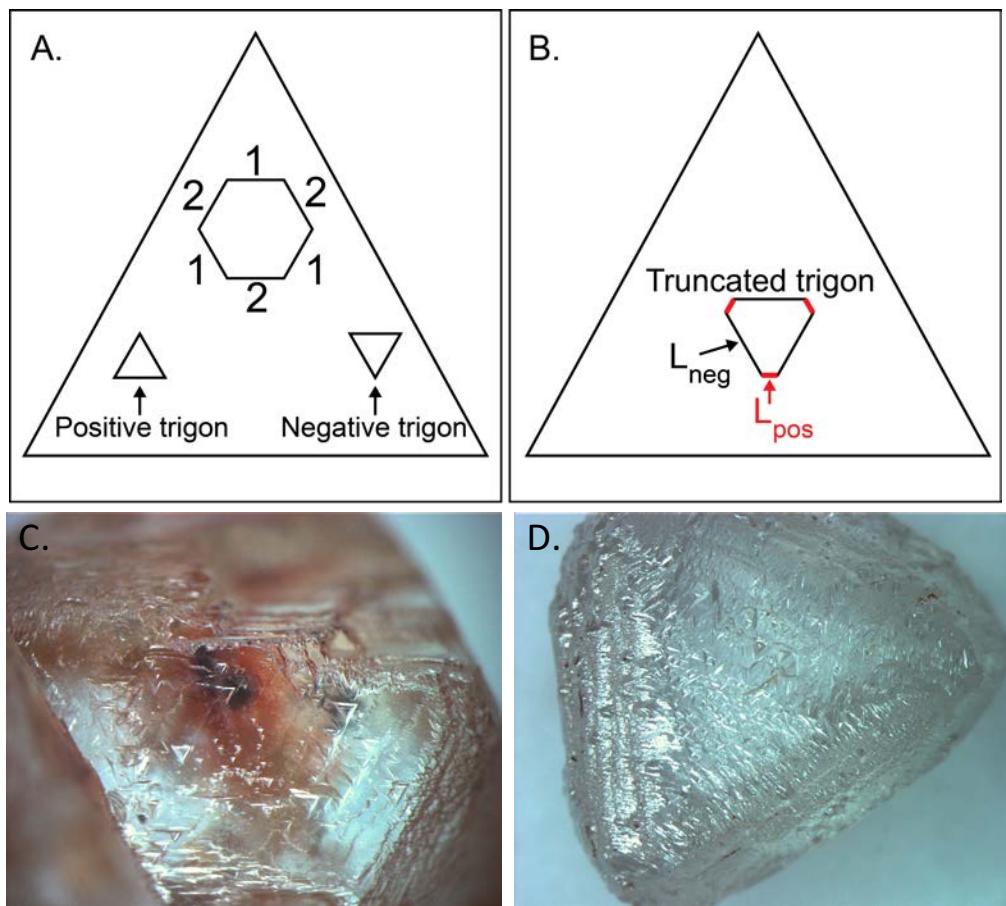
Figure 1
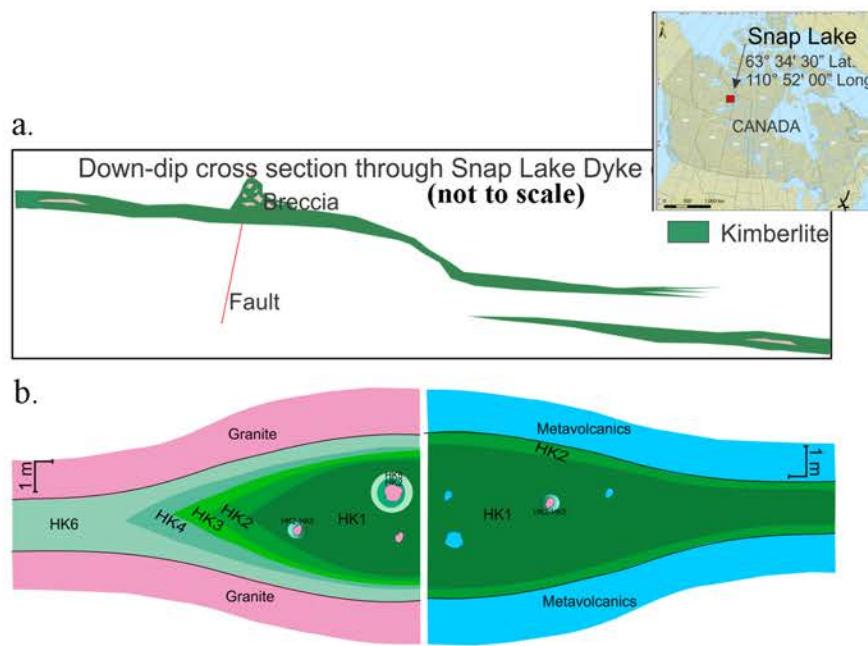
Figure 2

Wall type \ Bottom type	Flat-bottomed (f/b)	Point-bottomed (p/b)	Curved bottom (c/b)
V type	$\alpha_{\{111\}}$ diameter / depth	depth	Not observed
U type	α_1 α_2	β_1 β_2 Bottom type is determined by $\beta_1 + \beta_2$	depth
Y type	α_1 α_2		Not observed

Figure 3
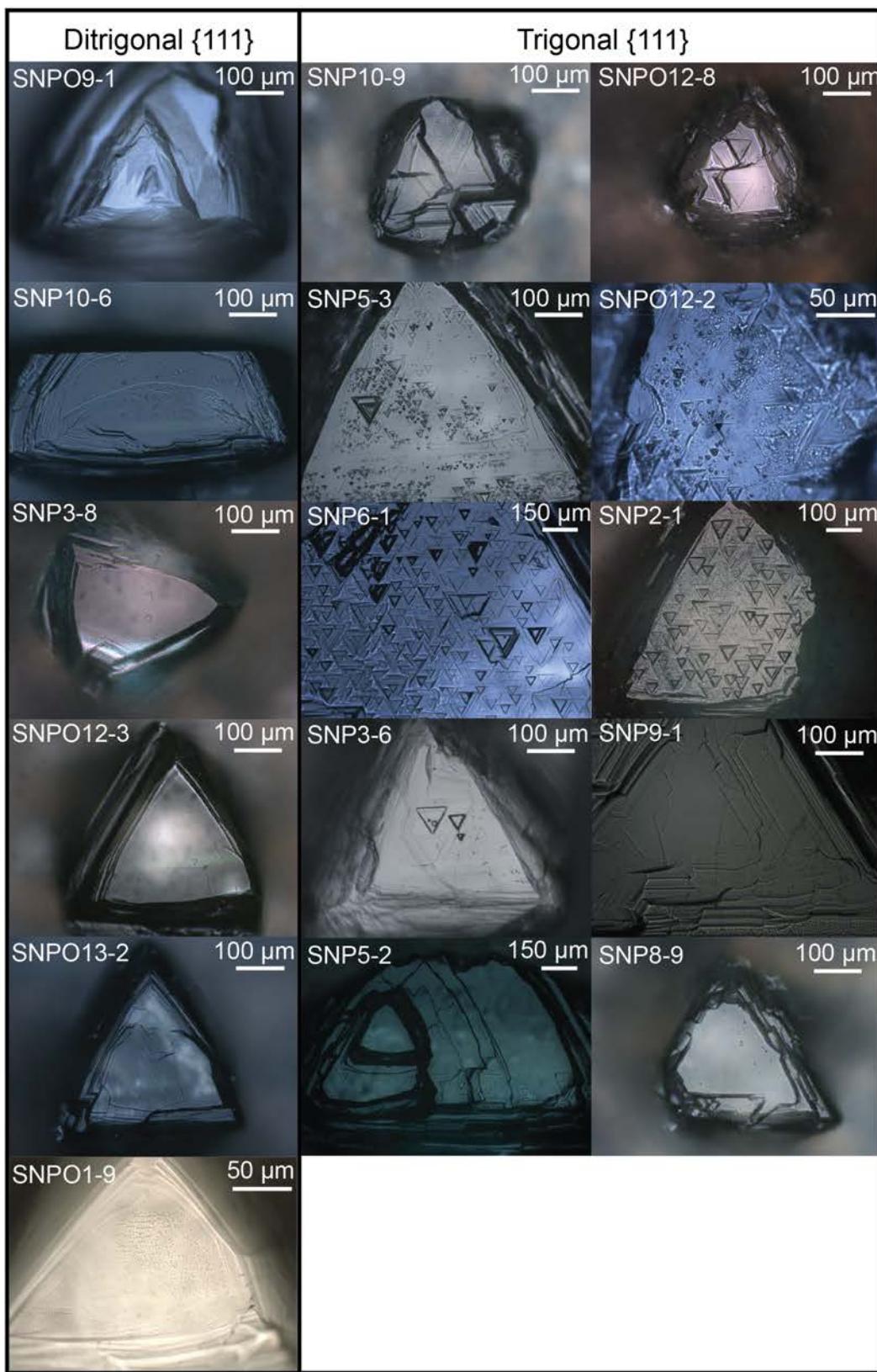
Figure 4

Snap Lake diamonds (this study)
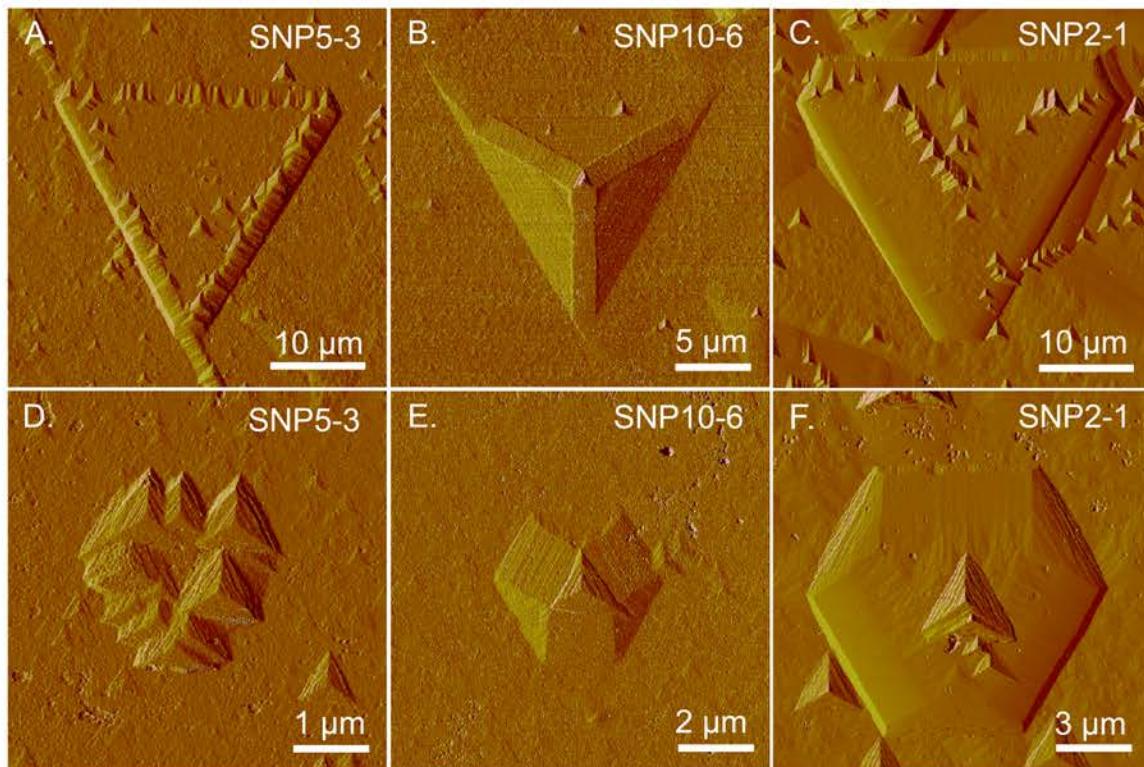
## Experiments in C-O-H fluids (after Zhang, 2016)
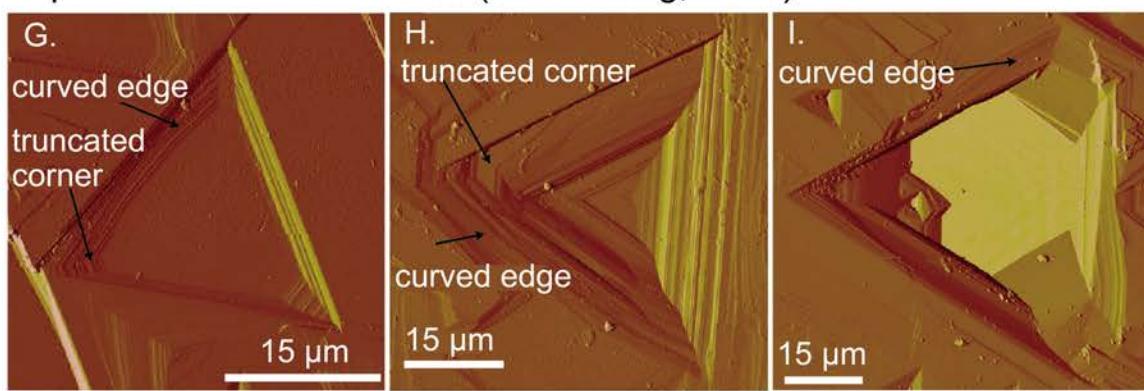
Figure 5
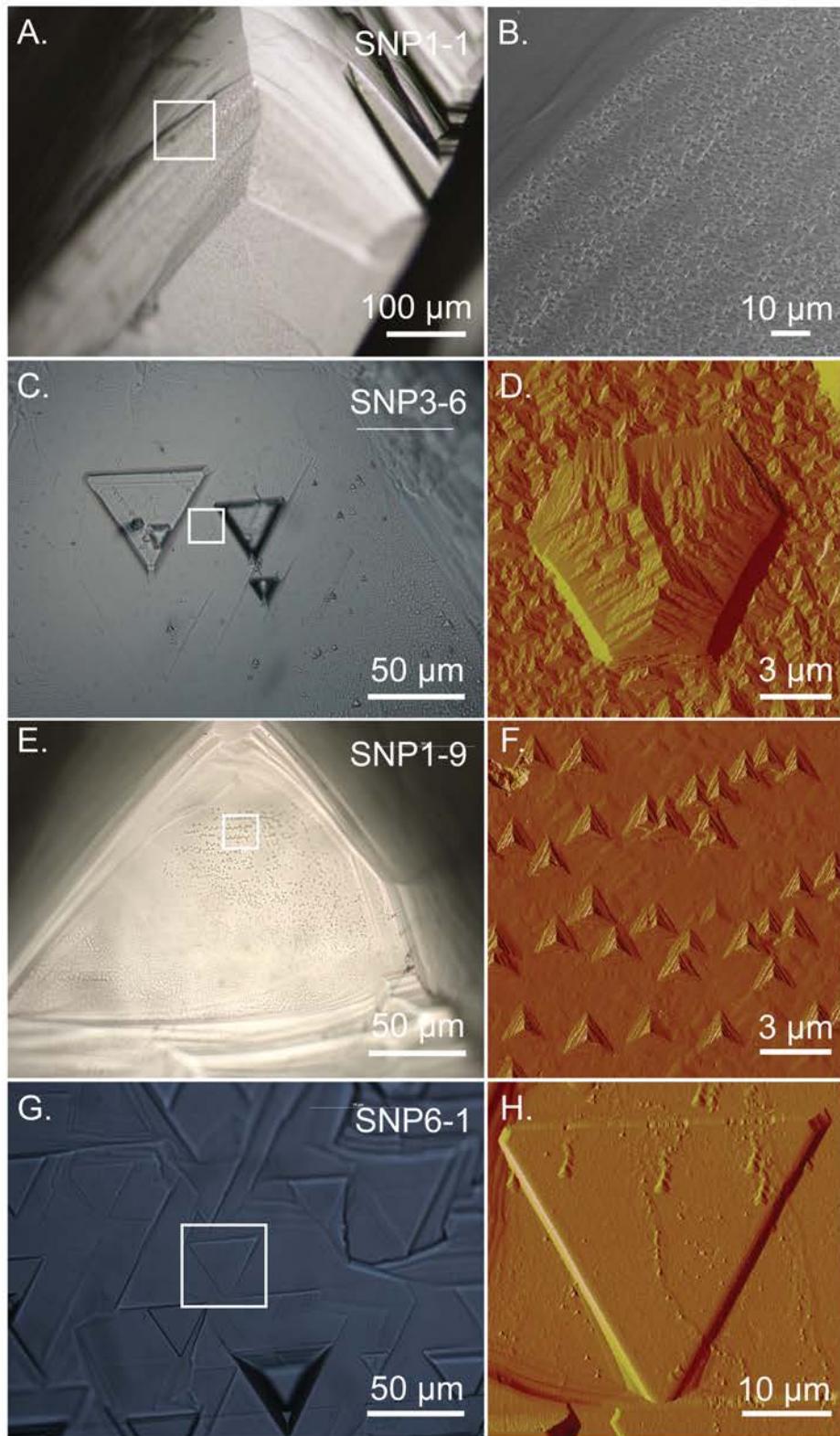
Figure 6
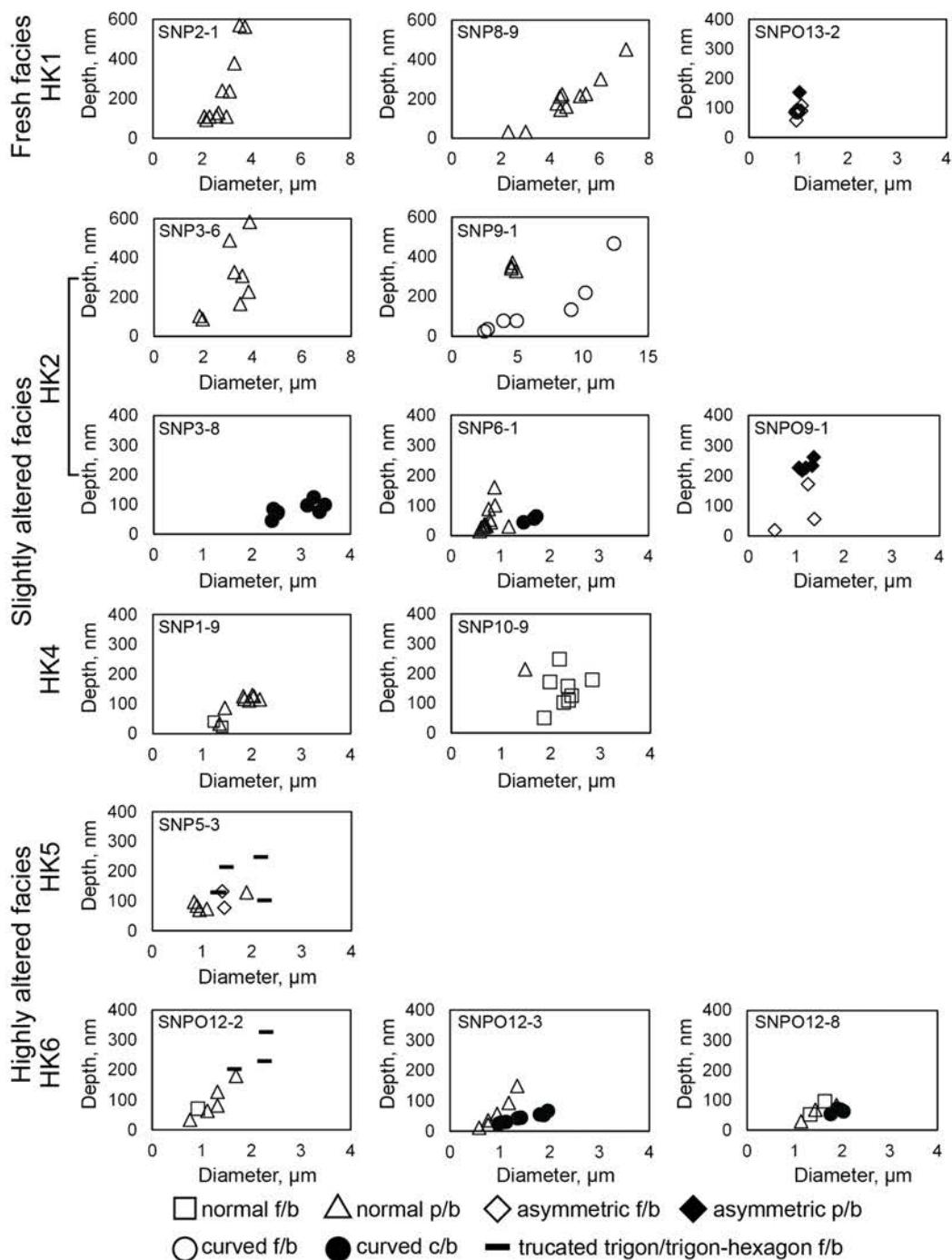
Figure 7
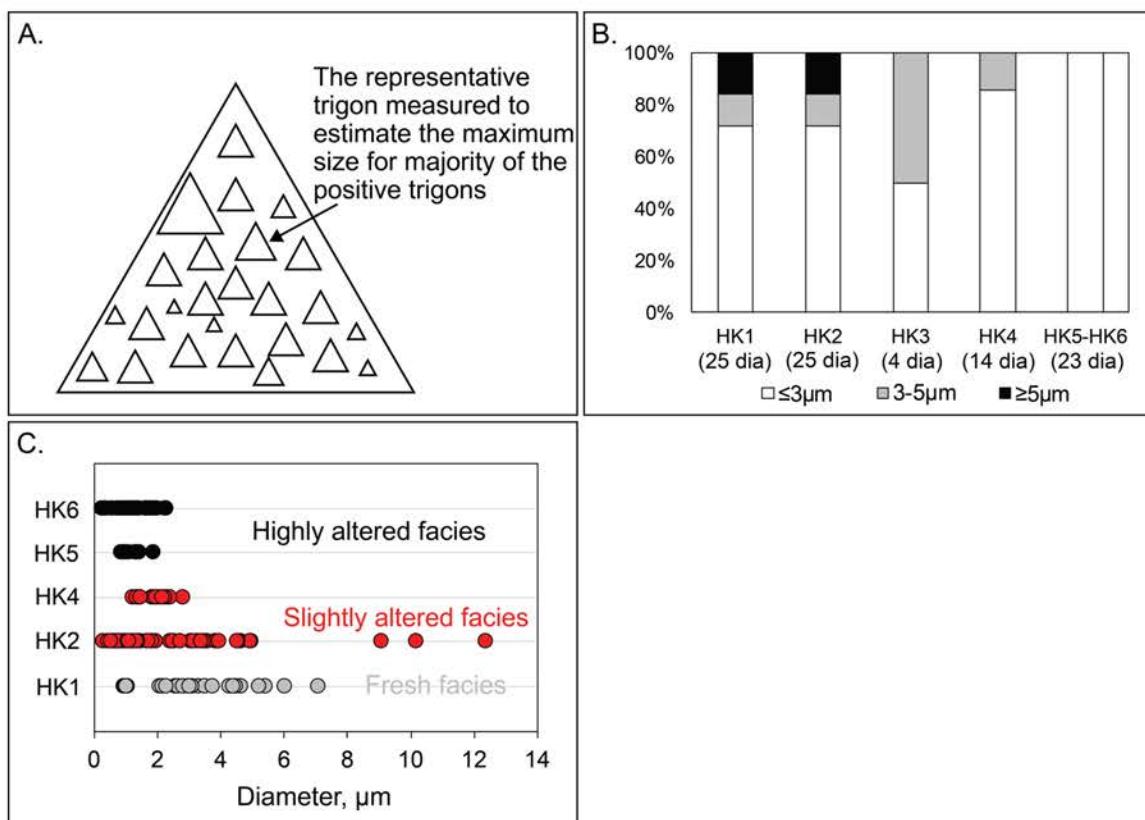
Figure 8
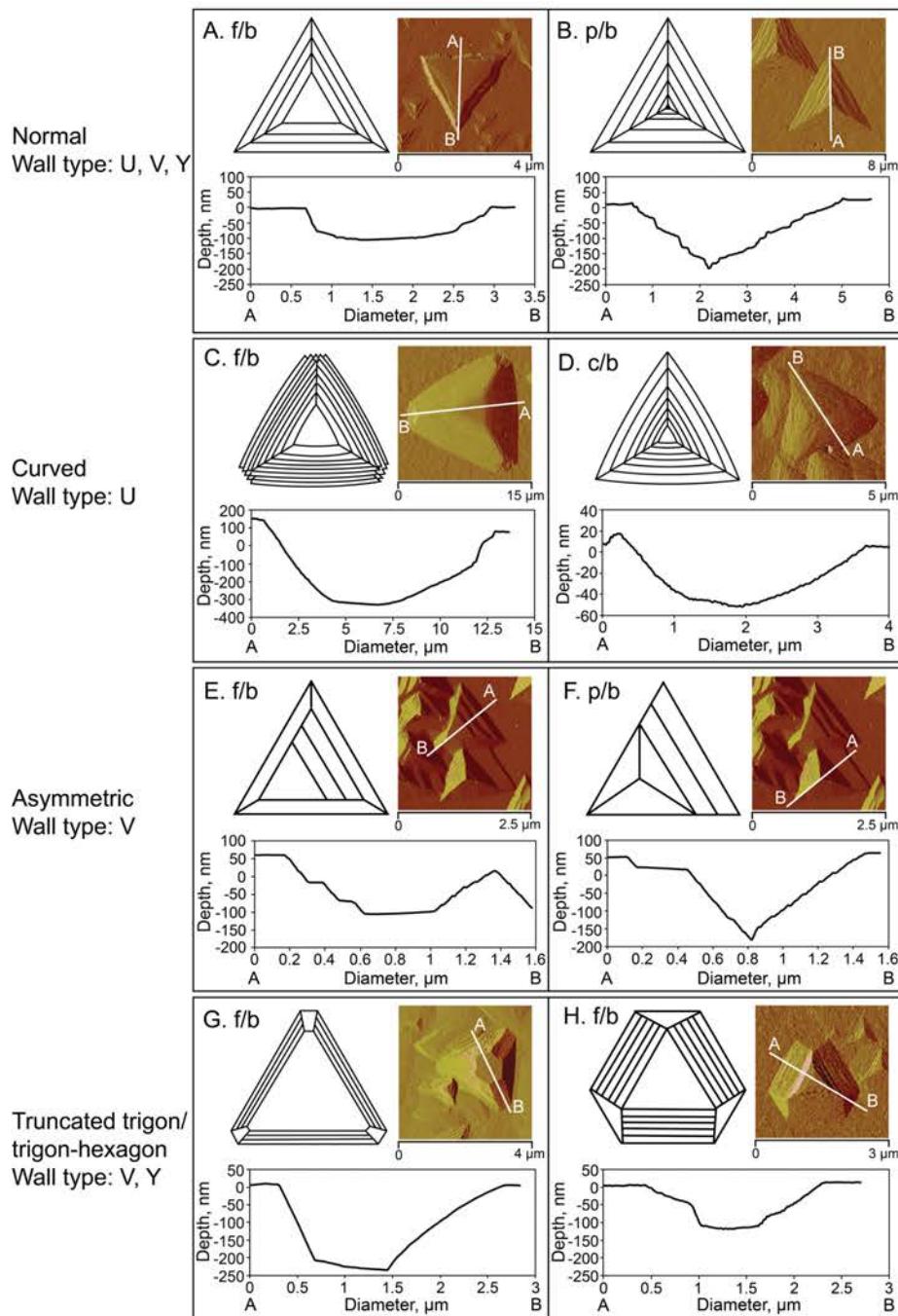
Figure 9
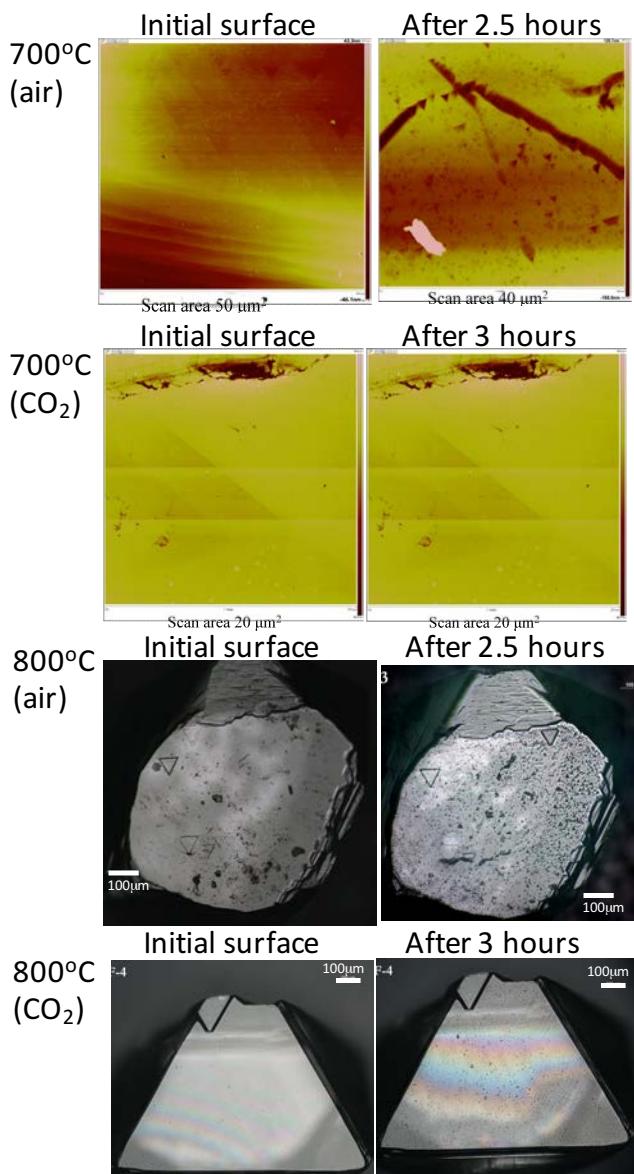
Figure 10

Figure 11

Figure 12