1	Revision 2
2	A new occurrence of corundum in eucrite and its significance
3	Jie-Ya Li ¹ , Ai-Cheng Zhang ^{1,2,*} , Naoya Sakamoto ³ , Hisayoshi Yurimoto ^{3,4,5} , Li-Xin Gu ⁶
4	¹ State Key Laboratory for Mineral Deposits Research, School of Earth Sciences and
5	Engineering, Nanjing University, Nanjing 210023 China
6	² CAS Center for Excellence in Comparative Planetology, China
7	³ Isotope Imaging Laboratory, Creative Research Institution, Hokkaido University,
8	Sapporo 010-0021, Japan
9	⁴ Department of Natural History Sciences, Hokkaido University, Sapporo 060-0810,
10	Japan
11	⁵ Institute of Space and Astronautical Science, Japan Aerospace Exploration Agency,
12	Kanagawa 252-5210, Japan
13	⁶ Institute of Geology and Geophysics, Chinese Academy of Sciences, Beijing 100029,
14	China
15	
16	*Corresponding author. Ai-Cheng Zhang, e-mail: aczhang@nju.edu.cn
17	Jie-Ya Li, e-mail: 1403219034@qq.com
18	Naoya Sakamoto, e-mail: naoya@ep.sci.hokudai.ac.jp
19	Hisayoshi Yurimoto, e-mail: yuri@ep.sci.hokudai.ac.jp
20	Li-Xin Gu, e-mail: gulixin@mail.iggcas.ac.cn
21	
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23	Abstract
24	Diversity of lithologies is an important proxy of internal evolution in differentiated
25	planets and asteroids. The major lithologies in Vesta, based on the howardite-eucrite-
26	diogenite clan meteorites, include basalt, gabbro, noritic orthopyroxenite,
27	orthopyroxenite, dunite, harzburgite, and dacite. No other lithology has been reported
28	up to date. In this study, we report a new occurrence of corundum in eucrite meteorite
29	Northwest Africa (NWA) 8647. Three-dimensional petrographic observations reveal
30	that the corundum grain occurs as a mineral inclusion in highly deformed pyroxene
31	fragment. The texture indicates that the corundum is not a contaminant. The
32	corundum-associated pyroxenes have Fe-Mn compositions consistent with typical
33	pyroxenes from howardite-eucrite-diogenite meteorites. We suggest that the
34	corundum grain could be an xenocryst incorporated during the ascent of a basaltic
35	magma. The results might indicate the presence of an Al-rich, Si-poor region,
36	probably lithology in the interior of Vesta, implying that the evolution and internal
37	structure should be much more complex than previously thought.
38	Keywords: corundum; Northwest Africa 8647, Al-rich lithology; eucrite; Vesta

INTRODUCTION

40 The magma ocean concept is one of the most fundamental hypotheses in Earth and planetary Sciences. In magma ocean models, differentiated celestial bodies 41 42 have experienced a global melting and differentiation after accretion. The magma ocean 43 phase has been considered as the starting point for most of the differentiated 44 planets and asteroids (Elkins-Tanton, 2012). However, records of the magma ocean 45 phase in the terrestrial planets (such as Earth and Mars) are extremely rarely preserved due to later complex processes including plate tectonics and weathering. 46 47One of the methods of constraining the earliest evolution of the terrestrial planets may 48 rely on the investigations of differentiated protoplanets, in which the records of early 49 stage evolution have been largely preserved (Zuber et al., 2011).

50 Asteroid 4 Vesta is the largest known protoplanet in our solar system with a 51 core-mantle-crust structure (Russell et al., 2012) and its differentiation might have taken 52 place a few million years after the formation of the earliest solid material in the solar 53 system (Wadhwa et al., 2009). Therefore, Vesta plays an important role in 54 understanding early processes and evolution of terrestrial planets. Meanwhile, the 55 differentiation and evolution of Vesta can be constrained based on the diversity and 56 origin of lithologies found in meteorite samples from Vesta and the remote-sensing data 57of Vesta. In the past decades, many investigations have been performed on howardite-58 eucrite-diogenite (HED) meteorites, the samples that have likely been derived from 59 Vesta (Mittlefehldt, 2015). The lithologies that have been reported in HED meteorites 60 include basalt, gabbro, noritic orthopyroxenite, orthopyroxenite, dunite, harzburgite, and dacite (e.g., Beck and

McSween, 2010; McSween et al., 2011; Mittlefehldt, 2015; Hahn et al., 2017; Zhang et
al., 2020).

63 Corundum is an important mineral indicator of Al-rich compositions for terrestrial 64 rocks (Giuliani et al., 2014). In HED meteorites, it was only reported as a shock-induced 65 mineral in the eucrite Northwest Africa 8003 (Pang et al., 2018, 2019). Recently, during 66 investigation of shock-metamorphism in HED meteorites, a corundum grain was 67 observed as an inclusion in pyroxene in the Northwest Africa (NWA) 8647 eucrite. Its 68 occurrence is different from other extraterrestrial occurrences of corundum including that 69 was described in Pang et al. (2018, 2019). In this study, we report its occurrence, 70 identification, and potential significance for a hidden Al-rich, Si-poor region in the 71 interior of Vesta.

72

ANALYTICAL METHODS

73 The polished section of the NWA 8647 used in this study was prepared in the 74following sequence. First, a thin chip of NWA 8647 was cut with diamond blade and 75 attached on a 1-inch rounded silica slide with epoxy. Then, the sample was thinned to 76 approximately 0.1 mm in thickness with SiC abrasive papers. After that, the sample was 77 polished with diamond pastes of various particle sizes (5 μ m, 1 μ m, and 0.5 μ m). During 78 the whole process of sample preparation, no alumina polishing paste was used. Before 79 being observed with the scanning electron microscope (SEM), the sample was carbon 80 coated.

Petrographic textures in the meteorite NWA 8647 were observed using a Zeiss Supra 55 scanning electron microscope under backscattering electron (BSE) mode at Nanjing University, Nanjing, China. An energy dispersive spectrometer (EDS) installed

84 on this SEM instrument was used to qualitatively identify mineral phases in the polished 85 section. Quantitative mineral chemical analyses were obtained by wavelength dispersive 86 spectrometers installed in a JEOL 8100 electron probe microanalyzer (EPMA) at Nanjing 87 University. An accelerating voltage of 15 kV was used. The EPMA analysis with a beam 88 current of 20 nA was carried out for most minerals in this study. Measurement times are 89 20 s for peaks and 10 s for background, respectively. Natural and synthetic standards 90 were used for concentration calibration. All data were reduced with the ZAF (atomic 91 number-absorption-fluorescence) procedure.

92 The structural identification of corundum and surrounding minerals was performed 93 using an electron backscatter diffraction detector (EBSD) attached on the JEOL 7000F 94 field emission SEM instrument at Hokkaido University, Sapporo, Japan. Before the 95 EBSD analyses, the sample was vibro-polished with silica suspension and carbon coated. 96 The EBSD pattern and EDS pattern of minerals were obtained simultaneously with the 97 Aztec software. An accelerating voltage of 20 kV and a beam current of 4 nA were used. 98 The EBSD patterns of corundum were indexed with various polymorphs of Al_2O_3 . The 99 Aztec software automatically suggests indexing solutions ranked by the lowest "mean 100 angular deviation" (MAD) as an index of "goodness of fit." MAD numbers <1 are 101 considered desirable for accurate solutions (Ma and Rossman, 2008; Zhang et al., 2015).

To constrain the textural relationship between corundum and surrounding materials in three dimensions, an ultrathin section was cut through the corundum grain perpendicular to the thin section surface using the focused ion beam (FIB) technique at the Institute of Geology and Geophysics, Chinese Academy of Sciences, Beijing. The FIB milling was conducted on a Zeiss Auriga Compact SEM instrument. A focused Ga107 ion beam which was accelerated under various high voltages (4–30 kV) was used 108 to sputter material from the sample. Before fine milling, a few secondary electron images 109 of the ultrathin section were made at an accelerating voltage of 1.5–5 kV. After that, 110 the ultrathin section was mapped with SEM-EDS at a 10-kV accelerating voltage. The 111 final section is approximately 100 nm in thickness. Unfortunately, the ultrathin 112 section was lost from the Cu grid before observation with transmission electron 113 microscope.

114

RESULTS

115 The NWA 8647 meteorite is a 326-gram brecciated eucrite (Bouvier et al., 2017). 116 The polished section of NWA 8647 used in this study contains two petrographically 117 different portions (Fig. 1). In one portion, the sample was largely melted and 118 recrystallized. However, lithic fragments and pyroxene, plagioclase, and silica mineral 119 fragments are commonly observed in this portion. In the fine-grained, melted 120 and recrystallized region, pyroxene and plagioclase form a fine-grained, subophitic 121 texture, in which pyroxene grains have a large chemical variation $(En_{12,9-59,9}Fs_{31,9-5})$ 122_{61.2}Wo_{8.3-26.7}) with a Fe/Mn value of 23-32 (Table S1, Figs. 2-3). The composition of 123 plagioclase is An_{87,2-91,3}Ab_{8,4-12,6}Or_{0,2-0,3} (Table S2).

The other portion of the polished section was not melted and has a complex petrographic texture (Fig. 1). A few shock-induced melt veins are present in this unmelted portion (Fig. 1). The shock-induced melt veins are composed mainly of fine-grained clinopyroxene grains with cation vacancies (compositions are not given in the present paper). No other high-pressure phases have been observed in this meteorite. Some unmelted regions show a coarse-grained, ophitic-subophitic texture of pyroxene and plagioclase, in which pyroxene usually contains exsolution lamellae

(up to 25 µm in

130 width, Fig. 4). Both low-Ca pyroxene and high-Ca pyroxene with exsolution texture have limited compositional variations, En36.3-39.2Fs57.3-61.1Wo1.8-4.4 and En29.4-30.9Fs25.1-131 132 27.8W042.2-44.6, respectively (Fig. 2). The Fe-Mn compositions of the coarse-grained 133 pyroxenes (especially low-Ca pyroxene) plot along the trend line for HED meteorites 134 (Fig. 3; Papike et al., 2003). The mean compositions of the coarse-grained low-Ca 135 pyroxene and high-Ca pyroxene with exsolution texture are En_{37.7}Fs_{59.5}Wo_{2.8} and 136 $En_{30.0}Fs_{26.6}Wo_{43.4}$, respectively, suggesting a two-pyroxene equilibrium temperature at 137 816 °C with the assumption of a pressure at 10 kbar (Brey and Köhler, 1990). The 138 plagioclase compositions are An_{87,4-90,5}Ab_{9,3-12,2}Or_{0,2-0,5} (Table S2).

139 The corundum grain in NWA 8647 is observed in a large pyroxene fragment (~280 140 µm in size), which is located in the fine-grained melted and recrystallized portion of NWA 8647 (Figs. 1 and 5a). This pyroxene fragment has been largely affected by shock 141 142 metamorphism. The major part of the pyroxene fragment has deformed and transformed into fine-grained pyroxene grains approximately 1-2 µm size. 143 However, high-Ca pyroxene exsolution lamellae (En_{33,7-35,0}Fs_{26,3-29,2}Wo_{36,3-40,0}) can 144 be still recognized within low-Ca pyroxene (En_{40.8-43.5}Fs_{47.8-54.9}Wo_{2.7-10.9}). The Fe-Mn 145 compositions of the pyroxenes plot on the trend line for HED meteorites (Fig. 3). 146 147 However, in the pyroxene quadrilateral diagram, the pyroxenes associated with the 148 corundum grain plot slightly to the Mg-rich side of the coarse-grained pyroxenes in 149 the unmelted regions (Fig. 2). Moreover, the low-Ca pyroxene in the fragment 150 associated with corundum is also slightly Ca-enriched compared to the coarse-grained 151 low-Ca pyroxene in unmelted portion; whereas the high-Ca pyroxene in the fragment 152associated with corundum is also slightly Ca-depleted compared with the coarse-grained high-Ca pyroxene in the unmelted portion

153 (Fig. 2). Fine-grained polycrystalline chromite is also present as inclusions in the154 pyroxene fragment (Fig. 5b).

155 The corundum grain is triangular in shape and approximately 4 µm in its largest 156 dimension (Fig. 5c). The EPMA results reveal it contains dominant Al₂O₃ (98.1–98.5 157 wt%) with minor SiO₂ (0.53-0.55 wt%) and FeO (1.15-1.18 wt%), which might be due 158 to beam-overlapping on surrounding pyroxene. The corundum grain is in tight contact 159 with the host pyroxene grain in the thin-section plane and the plane perpendicular to the 160 thin-section surface (Figs. 5c and 6). Based on the high-magnification SEM image of the 161 FIB section, a few tiny, anhedral plagioclase grains ($<0.6 \mu m$ in length) are associated 162 with the pyroxene (Fig. 6); however they are not in direct contact with the corundum 163 grain. Structure of the corundum grain is confirmed based on its EBSD pattern, which is 164 best indexed with the $R\bar{3}c$ corundum structure (MAD=0.21; Fig. 7) and cannot be 165 indexed with other Al₂O₃ phases. The crystal structure of the pyroxene fragment 166 including the corundum grain was also confirmed with EBSD patterns.

167 To understand a potential reason why the pyroxene fragment including corundum 168 has compositions different from the coarse-grained pyroxene in the unmelted portion, 169 compositions of polycrystalline pyroxenes in a large lithic fragment (Fig. 8), also located 170 in the melted and recrystallized portion, were also measured with EPMA (Table S1). The 171polycrystalline pyroxenes have a grain size varying from 5 to 10 µm and show Z-contrast 172 heterogeneity in the BSE image. Some relatively large grains show a bright-dark-bright 173 zoning texture from the core to the rim (Fig. 8b). The rims of pyroxene grains were not 174 measured to avoid potential overlapping on surrounding plagioclase. At the interface 175between pyroxene and plagioclase, plagioclase usually occurs as thin lath-like crystals (Fig. 8b). Measurements on the pyroxene grains show a Fe-Mn compositional variation consistent with the trend line for HED meteorites (Fig. 3), while these pyroxenes plot with a large variation slightly to the Mg-rich side of the coarse-grained pyroxenes from the unmelted portion, but to the Fe-rich side of the pyroxene fragment including corundum in the pyroxene quadrilateral diagram (Fig. 2).

181

DISCUSSION

182 Corundum is usually used as a polishing material for sample preparation in many 183 laboratories. In addition, corundum can exist in many terrestrial rocks. Therefore, we are 184 very cautious about the source of the corundum grain in our sample. During the whole 185 sample preparation processes in our laboratory, no alumina powder was used. Only SiC 186 and diamond were used as polishing materials. This would largely decrease the 187 possibility that the corundum grain was a contaminant during sample preparation. 188 Importantly, the corundum grain is in tight contact with the host pyroxene in three 189 dimensions based on the observations on the thin-section plane and the FIB-section plane. 190 This feature is different from the occurrences of polishing paste contaminants in the 191 literature (e.g., Dobrzhintskaya et al., 2014). Therefore, the possibility that the corundum 192 grain in NWA 8647 was contamination during sample preparation can be excluded.

193 Corundum is also an important phase in chondritic meteorites, as presolar grains 194 (e.g., Takigawa et al., 2018 and references therein) and as a primary or secondary phase 195 in Ca,Al-rich inclusions (e.g., Simon et al., 2002; Ma et al., 2009; Makide et al., 2009, 196 2013 and references therein). Exotic chondritic fragments have usually been observed in 197 brecciated HED meteorites (e.g., Zolensky et al., 1996; Lorenz et al., 2007; and 198 references therein). This imposes a possibility that the corundum grain and its associated

199 pyroxenes were derived from other asteroids rather than the eucrite parent body. However, 200 the occurrence of corundum in NWA 8647 is largely distinct from those of chondritic 201 corundum. For instance, presolar grains in chondrites are much smaller than the 202 corundum grain and usually occur as discrete grains in the fine-grained matrix in 203 chondrites (Zinner, 2014). Therefore, it is reasonable to consider that presolar corundum 204 grains might also occur mainly as discrete grains. The primary and secondary corundum 205 grains in Ca,Al-rich inclusions are usually associated with other primary or secondary 206 Ca,Al-rich minerals (e.g., melilite, grossular, nepheline; Simon et al., 2002; Ma et al., 207 2009; Makide et al., 2009, 2013 and references therein). However, in the current study, 208 corundum appears totally enclosed in pyroxenes in three dimensions. Furthermore, the 209 corundum-associated pyroxene grains in NWA 8647 show an exsolution texture, which is 210 absent in chondritic materials. Importantly, the corundum-associated pyroxene fragment 211 in NWA 8647 have a Fe-Mn compositional variation well consistent with typical HED 212 pyroxenes (Fig. 3). Therefore, the corundum grain and its associated pyroxenes are not 213 exotic chondritic fragment. Instead, they were probably derived from the eucrite parent 214 body.

The major element compositions of the corundum-associated pyroxene fragment in NWA 8647 are slightly different from those of the coarse-grained pyroxenes in the unmelted portion (Fig. 2). This deviation can be explained if the corundum-associated pyroxenes had a source different from the other pyroxenes in NWA 8647. Alternatively, the compositional deviation might be due to elemental exchange with the surrounding melt during and after the melting and recrystallization of the sample. Similar textural and chemical features have been observed for olivine and chromite in shocked lunar 222 meteorites and some shocked eucrites (e.g., Zhang et al., 2011; Pang et al., 2017). During 223 the shock-induced melting, the pyroxene fragment was highly deformed and many 224 pyroxene grains approximately $1-2 \mu m$ in size formed. The small pyroxene sub-grains 225 might have exchanged Mg, Fe, and Ca with surrounding melt. During shock-induced high temperature and fast cooling, Mg would be preferably incorporated into 226 227 pyroxene structure, which can account for why the compositions of the pyroxene fragment including corundum plot to the Mg-rich side of the coarse-grained 228 229 pyroxenes in the unmelted portion (Fig. 2). Presence of the tiny plagioclase grains 230 shown in Fig. 6 might indicate that high-temperature melt migrated into the grain 231 boundaries between fine-grained pyroxenes in the fragment, although such tiny 232 grains cannot be recognized at low-magnification BSE images. Such an 233 interpretation is generally supported by the compositional deviation of the 234 polycrystalline pyroxenes in the lithic fragment shown in Fig. 8 from the coarse-grained pyroxenes in the unmelted portion (Fig. 2). The pyroxene grains shown in Fig. 8b have a 235 grain size of $5-10 \mu m$, larger than the pyroxene grains in the fragment including 236 237 corundum. The large grain size might only allow for limited elemental exchange, 238 which can account for why the pyroxene grains shown in Fig. 8b plot only slightly to 239 the Mg-rich size of the coarse-grained pyroxene in the unmelted portion (Fig. 2).

Corundum has been observed in the eucrite NWA 8003 (Pang et al., 2018, 2019). The corundum in NWA 8003 occurs as micron to submicron euhedral grains associated with ilmenite, vacancy-rich clinopyroxene, vestaite, and kyanite only in shock-induced melt pockets, which are usually surrounded by tissintite (Pang et al., 2019). No corundum grains were observed in the host rock of NWA 8003. Pang et al. (2019) interpreted the

245 corundum in NWA 8003 to be one of the crystallization products after partial melting 246 involving ilmenite, pyroxene, plagioclase, and silica under high pressures. Recently, Yang et al. (2019) described the presence of corundum in the ungrouped achondrite 247 NWA 7325. The corundum in NWA 7325 occurs as needle-like crystals (<0.9 µm in 248 length) in the interior of large plagioclase (Yang et al., 2019). The authors suggested that 249 250 the corundum might have crystallized from molten plagioclase, which can be accounted for by the melting reaction of anorthite at high temperature (Goldsmith, 251252 1980). In the present study, although the corundum-associated pyroxenes are 253 polycrystalline in texture, probably due to shock metamorphism and its heating effect, no melting reaction similar to those in NWA 8003 and NWA 7325 was observed in 254 255NWA 8647. The corundum-associated mineral assemblage in NWA 8647 is distinctly different from those of shock-induced corundum in NWA 8003. Although minor 256 plagioclase grains have been observed in the pyroxene fragment including corundum in 257 258 NWA 8647, the grain sizes of these plagioclase grains are much smaller than the 259corundum. Corundum was also not observed in other regions in NWA 8647. Based on the 260 current observations, the corundum grain in NWA 8647 cannot be a product of shock 261 metamorphism in the host rock. Instead, the corundum grain in NWA 8647 is most 262 likely be of indigenous origin.

Our observations reveal that the corundum grain in the current study could be a mineral inclusion in orthopyroxene with augite exsolution lamellae. During shock metamorphism of NWA 8647, deformation and polycrystallization took place in the pyroxenes, with the corundum grain not heavily affected. Given that eucritic melts are not corundum-normative (Stolper, 1975), the corundum grain in NWA 8647 was probably captured as an xenocryst during the ascent of a basaltic magma and

included by a

pyroxene grain that has crystallized from the basaltic melt. This origin might be similar to
the corundum in terrestrial alkali basaltic extrusions (e.g., Guo et al., 1996; Sutherland et
al., 1998).

271Natural corundum has been widely observed in a variety of terrestrial lithologies 272 (e.g., Giuliani et al., 2014) and primitive chondrites (e.g., Simon et al., 2002; Makide et 273 al., 2013; Takigawa et al., 2018). In terrestrial magmatic rocks, it was observed from 274 syenites and nepheline syenites (e.g., Moyd, 1949), to quartz-free pegmatites (e.g., Rao et 275al., 2012), porphyry copper deposits (e.g., Bottril, 1998), to alkali basaltic extrusions (e.g., 276 Guo et al., 1996; Sutherland et al., 1998), alkaline basic lamprophyre (e.g., Brownlow 277 and Komorowski, 1988), and kimberlites (e.g., Mazzone and Haggerty, 1989). In the 278 latter three types of occurrence, corundum occurs usually as xenocrysts or in xenoliths. In 279 addition, corundum has also been observed in a variety of high-grade metamorphic 280 aluminous rocks (c.f., Giuliani et al., 2014). In primitive chondrites, corundum grains 281 occur either as presolar grains in fine-grained matrix (e.g., Takigawa et al., 2018) or as a 282 component in Ca-Al-rich inclusions (e.g., Simon et al., 2002; Makide et al., 2013). 283 Although these natural rocks and objects containing corundum vary largely in lithology, 284 their common characteristic is Al-rich and Si-poor, at least locally (Giuliani et al., 2014). 285 Therefore, the occurrence of corundum in NWA 8647 might indicate the presence of an Al-rich and Si-poor region, probably lithology locally in the interior of Vesta, although 286 287 the exact location of the Al-rich and Si-poor region/lithology remains unconstrained 288 based on the current observations.

289

IMPLICATIONS

290	No Al-rich and Si-poor rocks containing corundum have been identified and
291	considered in previous investigations on HED meteorites (c.f., McSween et al., 2011;
292	Mittlefehldt, 2015) and theoretical simulations (Righter and Drake, 1997; Ruzicka et al.,
293	1997; Mandler and Elkins-Tanton, 2013). The finding of a hidden Al-rich and Si-poor
294	region based on the presence of the indigenous corundum in NWA 8647 indicates that the
295	internal evolution of Vesta is more complex than previously thought. In future, more
296	detailed petrographic and mineralogical observations on HED meteorites are needed to
297	further understand the internal evolution and crystallization history of Vesta.
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431 Figure 1. Mosaic backscattered electron image of the section of NWA 8647. The left side

432 shown in this image was largely melted and recrystallized. However, the right side was

433 unmelted but contains shock-induced melt veins.



Figure 2. Pyroxene quadrilateral diagram showing the compositional variation of pyroxenes in NWA 8647. Blue circles represent compositions of the coarse-grained pyroxenes in the unmelted portion. Green diamonds represent compositions of the polycrystalline pyroxenes in the lithic fragment shown in Fig. 8. Red squares are compositions of the pyroxenes in the fragment hosting the corundum grain. Yellow squares represent compositions of fine-grained pyroxenes in the melted and recrystallized region.



Figure 3. Fe and Mn compositions of pyroxene in NWA 8647. The trend lines of
HED and Moon are adopted from Papike et al. (2003). The symbols in this figure are
same as those in Fig. 2.



447 Figure 4. Typical area from the unmelted region in NWA 8647 showing the exsolution

448 texture of pyroxene. Opx: orthopyroxene; Aug: augite; Pl: plagioclase.



Figure 5. BSE image of the pyroxene grain containing corundum (a) and the zoom-in images (b–c) with various magnifications of the local regions containing the corundum grain. In (b), two fine-grained chromite grains are also present and the upper one shows a polycrystalline texture. Pgt: pigeonite; Aug: augite; Opx: orthopyroxene; Pl: plagioclase; Crn: corundum; Chr: chromite.



- 456 Figure 6. Secondary electron image of the FIB section showing the relationship between
- 457 corundum and surrounding silicate minerals. Crn: corundum; Opx: orthopyroxene; Pl:
- 458 plagioclase; Pt: Pt metal.



460 Figure 7. EBSD pattern (a) of the labelled corundum crystal in Fig. 5b and the pattern (b)

461 indexed with the trigonal $R\overline{3}c$ structure (MAD=0.21).



Figure 8. (a) Backscattered electron image of relict lithic fragment in the melted region of
NWA 8647. The white box indicates the location of (b). (b) zoomed-in image of a typical
region from the upper part of (a) showing polycrystalline pyroxene. Note that plagioclase
grains in (b) have a high length/width ratio, indicating rapid crystallization from melt. Px:
pyroxene; Pl: plagioclase.