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1	Evolution of metamorphic fluid recorded in granulite facies metacarbonate
2	rocks from the middle segment of the Mogok metamorphic belt in central
3	Myanmar
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15	Short title: FLUID COMPOSITIONS OF MOGOK GRANULITES

#### 16 ABSTRACT

17 The Mogok metamorphic belt of Paleogene age, which records subduction- and 18 collision-related events between the Indian and Eurasian plates, lies along the western 19 margin of the Shan plateau in central Myanmar and continues northwards to the eastern 20 Himalayan syntaxis. Reaction textures of clinohumite- and scapolite-bearing 21 assemblages in Mogok granulite facies metacarbonate rocks provide insights into the 22 drastic change in fluid composition during exhumation of the collision zone. 23 Characteristic high-grade assemblages of marble and calc-silicate rock are clinohumite 24 + forsterite + spinel + phlogopite + pargasite/edenite + calcite + dolomite and scapolite 25 + diopside + anorthite + quartz + calcite, respectively. Calculated petrogenetic grids in 26 CaO-MgO-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub>-H<sub>2</sub>O-CO<sub>2</sub> and subsets of this system were employed to deduce 27 the pressure-temperature-fluid evolution of the clinohumite- and scapolite-bearing 28 assemblages. These assemblages suggest higher temperature (> 780–810 °C) and  $X_{CO2}$ 29  $[= CO_2/(CO_2 + H_2O) > 0.17-0.60]$  values in the metamorphic fluid for the peak 30 granulite facies stage, assuming a pressure of 0.8 GPa. Calcite grains commonly show 31 exsolution textures with dolomite particles, and their reintegrated compositions yield 32 temperatures of 720-880 °C. Retrograde reactions are mainly characterized by a 33 reaction zone consisting of a dolomite layer and a symplectitic aggregate of tremolite 34 and dolomite grown between clinohumite and calcite in marble, and a replacement 35 texture of scapolite by clinozoisite in calc-silicate rock. These textures indicate that the 36 retrograde reactions developed under lower temperature (< 620 °C) and  $X_{CO2}$  (< 37 0.08–0.16) conditions, assuming a pressure of 0.5 GPa. 38 The metacarbonate rocks share metamorphic temperatures similar to the Mogok 39 paragneiss at the peak granulite facies stage. The  $X_{CO2}$  values of the metacarbonate rock

40	at peak metamorphic stage are, however, distinctly higher than those previously
41	deduced from carbonate mineral-free paragneiss. Primary clinohumite, phlogopite, and
42	pargasite/edenite in marble have F-rich compositions, and scapolite in calc-silicate rock
43	contains Cl, suggesting a contrast in the halogen compositions of the metamorphic
44	fluids between these two lithologies. The metamorphic fluid compositions were
45	probably buffered within each lithology, and the effective migration of metamorphic
46	fluid, which would have extensively changed the fluid compositions, did not occur
47	during the prograde granulite facies stage throughout the Mogok metamorphic belt. The
48	lower $X_{CO2}$ conditions of the Mogok metacarbonate rocks during the retrograde stage
49	distinctly contrast with higher $X_{CO2}$ conditions recorded in metacarbonate rocks from
50	other metamorphic belts of granulite facies. The characteristic low $X_{CO2}$ conditions were
51	probably due to far-ranging infiltration of H <sub>2</sub> O-dominant fluid throughout the middle
52	segment of the Mogok metamorphic belt under low-amphibolite facies conditions
53	during the exhumation and hydration stage.
54	
55	KEYWORDS: clinohumite; scapolite; granulite facies; metamorphic fluid; Mogok
56	metamorphic belt

## 58 1 | INTRODUCTION

59 Metamorphic fluid effectively transports heat and elements during metamorphism, and 60 its chemical composition is one of the most important factors controlling the stability of mineral assemblages (e.g., Carmichael, 1970; Connolly & Trommsdorff, 1991; Ferry, 61 62 1994; Valley, 1986). In particular, metamorphosed carbonate rock typically yields 63 various compositions of CO<sub>2</sub>-bearing fluid by decarbonation and dehydration processes, 64 and infiltration of these fluid phases have significant effects on the mineral parageneses 65 of the surrounding lithologies by fluid-rock interaction. Studies of metacarbonate rocks 66 have proved useful in determining the pressure (P)-temperature (T)-fluid history of high-grade metamorphic regions (Buick, Cartwright & Williams, 1997; Dasgupta, 1993; 67 68 Faryad, 2002; Gallien, Mogessie, Bjerg, Delpino & de Machuca, 2009; Harley, 69 Fitzsimons & Buick, 1994; Motoyoshi, Thost & Hensen, 1991; Piazolo & Markl, 1999; 70 Satish-Kumar, Wada, Santish & Yoshida, 2001; Warren, Hensen & Ryburn, 1987). In 71 the case of granulite facies metamorphism, F- and Cl-substitutions of OH effectively 72 enlarge the stability fields of hydrous silicates, such as amphibole and biotite, towards 73 high temperature (e.g., Bose, Das & Fukuoka, 2005; Higashino, 2013; Motoyoshi & 74 Hensen, 2001; Pan & Fleet, 1996; Tsunogae et al., 2003). 75 Clinohumite is a typical mineral in metacarbonate rocks of granulite facies, and its F 76 concentration strongly controls the stabilities of clinohumite itself and, eventually, of 77 the mineral parageneses in silica-undersaturated systems (Rice, 1980). Scapolite, which 78 contains Cl and CO<sub>3</sub> as major components, is a main constituent phase of some 79 calc-silicate rocks, and its occurrence provides a strong constraint on metamorphic 80 temperature and fluid composition (e.g., Harley et al., 1994; Moecher & Essene, 1990; 81 Satish-Kumar & Harley, 1998). Evolution of metamorphic fluid compositions has been

82 discussed for metacarbonate rocks from many granulite facies regions: e.g., India (Satish-Kumar, Santosh, Harley & Yoshida, 1996; Satish-Kumar et al., 2001), Africa 83 84 (Kaneko, Miyano & Tsunogae, 2005), Antarctica (Satish-Kumar, Hermann, Tsunogae 85 & Osanai, 2006), and Australia (Buick et al., 1997). Most of these studies have 86 considered recrystallizations under extremely high  $X_{CO2}$  [= CO<sub>2</sub>/(CO<sub>2</sub> + H<sub>2</sub>O)] fluid 87 conditions at the granulite facies stage as well as the subsequent retrograde stage. 88 However, Buick, Cartwright and William (1997) reported that metacarbonates of the 89 Reynolds Range group (~0.5 GPa/750–800 °C) that initially equilibrated at  $X_{CO2} > 0.8$ 90 were mineralogically reset by local infiltration of H<sub>2</sub>O-rich fluids ( $X_{CO2} \le 0.02-0.3$ ) at 91 0.3-0.4 GPa/650-700 °C along narrow strike-parallel zones. 92 Granulite facies metamorphic rocks occur extensively in the middle segment of the

93 Late Eocene-Late Oligocene Mogok metamorphic belt in Myanmar, which formed 94 during subduction and collision between the Indian and Eurasian plates. Metacarbonate 95 rocks containing F-rich clinohumite or scapolite commonly occur in the granulite facies 96 area. These samples also preserve reaction textures of retrograde stages and provide 97 information for understanding the evolution of *P*-*T*-fluid conditions during exhumation. 98 The present study reports the compositional characteristics and modes of occurrence of 99 clinohumite, scapolite, and associated minerals in the metacarbonate rocks and 100 discusses changes of metamorphic fluid compositions from high- $X_{CO2}$  at the peak 101 metamorphic stage to low- $X_{CO2}$  during exhumation and a retrograde low-amphibolite 102 facies stage. These facts mean that firstly, the metamorphic fluid compositions were 103 internally buffered within each lithological unit and the effective migration of 104 metamorphic fluid did not occur during the peak granulite facies stage, and secondly, 105 their high  $X_{CO2}$  composition were extensively reset to low  $X_{CO2}$  during the retrograde

106	stage by infiltration of H <sub>2</sub> O-rich fluid phases beyond the lithologic boundaries
107	throughout the Mogok metamorphic belt. Abbreviations for the minerals and
108	endmembers described in the text, figures, and tables are after Whitney and Evans
109	(2010).
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111	
112	2   GEOLOGICAL SETTING
113	2.1 I Mogok metamorphic belt
114	Myanmar is geologically divided into Eastern and Western provinces, separated by the
115	1200 km long, N-S trending Sagaing dextral strike-slip fault (Figure 1). The Western
116	province, or Burma microplate, includes the Indo-Burma Ranges and Central Myanmar
117	basins. The Indo-Burma Ranges, where the Burma microplate overrides the Indian
118	oceanic crust, consist of Late Cretaceous to Paleogene marine sediments and
119	stratigraphically lower flysch-type sediments (Acharyya, 2007; Gardiner, Searle, Robb
120	& Morley, 2015; Ghose, Chatterjee & Fareeduddin, 2013). The Central Myanmar Basin
121	is covered by a series of Cretaceous to Pleistocene sediments and Quaternary volcanic
122	rocks (Mitchell, 1993). The Eastern province is a part of the Shan-Thai block that
123	collided with the Indochina block in the Early or Late Triassic (Metcalfe, 1988, 2000;
124	Mitchell, 1977). It comprises the Shan plateau, the Slate belt (or Mergui Group), and the
125	Mogok metamorphic belt (Figure 1). Most of the Shan plateau is characterized by an
126	Upper Cambrian to Lower Cretaceous succession (Boucot, 2002), overlying
127	metamorphic rocks of the Chaung Magyi Group. The Slate belt also consists of
128	distinctive Late Paleozoic glacial marine pebbly mudstones that extend to Thailand, the
129	Malay Peninsula, and Sumatra (Barber & Crow, 2009; Ridd & Watkinson, 2013).

130	The Mogok metamorphic belt extends along the northwestern margin of the Shan
131	plateau and west of the Slate belt, reaching 1500 km in length (Mitchell et al., 2007;
132	Searle et al., 2007). This belt consists of a series of undifferentiated high-grade
133	metasedimentary and meta-igneous rocks, and common lithologies include a variety of
134	gneiss, schist, quartzite, marble, calc-silicate rock, and migmatite with various granitoid
135	intrusions (Barley, Pickard, Khin Zaw, Rak & Doyle, 2003; Mitchell et al., 2007; Searle
136	et al., 2007). Radiometric dating focused on the metamorphic and tectonic evolution of
137	the Mogok belt and adjacent areas has been discussed by many researchers. Bertrand et
138	al. (1999) and Bertrand, Rangin, Maluski and Bellon (2001) reported Oligocene to
139	Middle Miocene ages based on Ar-Ar and K-Ar analyses of biotite and muscovite, and
140	proposed the northward passage of the eastern Himalayan syntaxis, characterized by
141	crustal thinning, resulting from the India-Asia oblique collision. Sensitive
142	high-resolution ion microprobe (SHRIMP) U-Pb dating of zircon obtained from granites
143	indicates that the magmatism occurred during the Jurassic and a later high-grade
144	metamorphic recrystallization event took place during the Eocene (Barley et al., 2003).
145	Garnier, Maluski, Giuliani, Ohnenstetter and Schwarz (2006) reported Ar-Ar ages of
146	18.7 – 17.1 Ma on phlogopite in ruby-bearing marble near Mogok. Searle et al. (2007)
147	reported U-Th-Pb ages of metamorphic monazite, zircon, xenotime, and thorite
148	employing isotope dilution thermal ionization mass spectrometry (ID-TIMS) and laser
149	ablation-multicollector-inductively coupled plasma mass spectrometry
150	(LA-MC-ICPMS), and suggested two distinct metamorphic events: firstly, a Paleocene
151	event, which implies earlier regional metamorphism (~59 Ma); and secondly, a Late
152	Eccene to Oligocene event, which was interpreted as the peak metamorphic stage
153	resulting in synmetamorphic crustal melting which produced garnet- and

154	tourmaline-bearing leucogranite (~24.5 Ma). Maw Maw Win, Enami and Kato (2016)
155	also dated the metamorphic monazite using U-Th-Total Pb Chemical Isochron Method
156	(CHIME: Montel et al., 2017 in press; Suzuki & Adachi, 1991) and reported the peak
157	metamorphic stage for upper-amphibolite and/or granulite facies as Late Eocene and the
158	subsequent hydration stage as Late Oligocene. The youngest magmatic event indicated
159	was Late Oligocene-Early Miocene, producing mantle-derived syntectonic hornblende
160	syenite and crust-derived leucogranite, postdating the high temperature metamorphism
161	and intrusion (Barley et al., 2003; Bertrand et al., 1999; Searle et al., 2007). These ages
162	suggest that the belt has experienced a complex series of tectonic, magmatic and
163	metamorphic events with subsequent post collisional extension, up-lift, and exhumation
164	processes.

166 2.2 | Onzon–Thabeikkyin region

167 The Onzon, Wabyudaung, Zayetkwin, Aung Tharyar, and Thabeikkyin areas, from168 which the samples were collected, are located about 100 km north of the Mandalay

169 region and belong to the middle segment of the Mogok metamorphic belt (Figure 2a).

170 These areas are denoted as the Onzon–Thabeikkyin region, hereafter, unless otherwise

171 noted. Paragneiss, marble, and calc-silicate rock occupy the middle segment of the

172 Mogok metamorphic belt from Mandalay to Mogok regions (Maw Maw Win et al.,

173 2016; Maw Maw Win, Enami, Kato & Ye Kyaw Thu, 2017; Ye Kyaw Thu, Enami,

174 Kato & Tsuboi, 2017; Ye Kyaw Thu, Maw Maw Win, Enami & Tsuboi, 2016;

175 Yonemura et al., 2013), continuing southward, where they become less dominant. The

176 metamorphic lithologies in this region form a zone with an average width of 40 km and

177 consist of various types of marble and calc-silicate rock with paragneiss of granulite

178 facies. The typical mineral assemblage of the paragneiss is Grt-Bt-Pl-Sil-Qz; cordierite 179 and spinel are included in some samples. Biotite grains coexisting with rutile and/or 180 ilmenite have a high TiO<sub>2</sub> content, of up to 6.9 wt% (Ye Kyaw Thu *et al.*, 2016). 181 Conventional geothermobarometers and Zr-in-rutile geothermometers yield *P*-*T* 182 conditions of 0.60–0.84 GPa/780–860 °C as the peak granulite facies metamorphism 183 (Ye Kyaw Thu et al., 2016, 2017). These paragneiss were extensively recrystallized 184 under lower-amphibolite facies conditions (about 0.40-0.55 GPa/620-640 °C) during 185 the exhumation and hydration stage. 186 In the Onzon–Thabeikkyin region, the metacarbonate rocks occur in a zone of 10–15 187 km width, which is wedged between paragneiss (Figure 2b). Myint Lwin Thein, Ohm 188 Myint, Sun Kyi and Hpone Myint Win (1990) divided these metamorphic rocks into the 189 Wabyudaung and Thabeikkyin Subgroups, which consist of Kantha, Tesoegyi, Sakantha, 190 and Kyetsaung Taung marble units and paragneiss and calc-silicate and diopside marble 191 units, respectively. The metacarbonate rocks widely occur as massive or thick-layered

192 forms and are locally intruded by granitoid rocks. The plutonic rocks are grouped into

193 biotite microgranite (Kabaing granite), leucogranite, syenite, and pegmatite. The

194 syenitic rocks occasionally include xenoliths of marble and calc-silicate rock, and their

195 U-Pb zircon ages yield ~30.9 Ma (Barley et al., 2003). The Kabaing granite, which is a

196 major plutonic lithology, intrudes into metamorphic rock units and parts of the syenite

197 and leucogranite, and shows no sign of tectonic deformation. U-Pb zircon and Ar-Ar

biotite ages of the Kabaing granite are  $16.8 \pm 0.3$  Ma (Gardiner *et al.*, 2016) and  $15.8 \pm$ 

199 1.1 Ma (Bertrand et al., 2001), respectively. Later-stage pegmatitic rocks locally intrude

200 into the Kabaing granite as dykes or veins, and grade into quartz vein or syenite

201 pegmatite.

202	Samples studied were collected from the Kyetsaung Taung marble unit of the
203	Wabyudaung Subgroup and the calc-silicate and diopside marble units and paragneiss
204	unit of the Thabeikkyin Subgroup (Figure 2b). The Kyetsaung Taung marble unit is
205	composed of major coarse-grained white marble consisting mostly of monomineralic
206	calcite grains and a small amount of impure marble containing calcite, diopside,
207	phlogopite, forsterite, humite, spinel, and graphite. This marble is massive and is
208	exposed as discontinuous bands trending ENE-WSW. Fine-grained marble and
209	graphite-marble are locally intercalated within this marble unit. The calc-silicate rock
210	unit forms a mappable zone, which can be traced continuously throughout the sampling
211	area, and is usually associated with a small amount of diopside marble, phlogopite
212	marble, and graphite marble. Thin quartzo-feldspathic bands with mica are locally
213	interbedded with the impure marble. Skarn is developed in the calc-silicate rock around
214	the Kabaing granite intrusion. Lenticular clots of graphite occur in metacarbonate rocks
215	adjacent to the boundary with the paragneiss unit. The paragneiss unit in the
216	Thabeikkyin area is mainly composed of highly weathered garnet-biotite paragneiss
217	with occasional intercalations of biotite paragneiss and leucogneiss.
218	
219	<b>3   PETROGRAPHY OF METACARBONATE ROCKS</b>
220	Four samples of dolomite-bearing marble (KST03, ZYK01, OZ01, and ACT01) and

three samples of calc-silicate rock (OZ03, WBY01, and KB01) were selected for

222 detailed analyses (Figure 2b). The mineral assemblages of the marble and calc-silicate

rock samples are listed in Table 1.

224

# 225 **3.1** | Forsterite-bearing marble

226	The forsterite-bearing marble samples, KST03 (96°04'35" E, 22°50'22" N) and ZYK01
227	(96°03'54" E, 22°49'06" N), were collected from the Kyetsaung Taung marble unit in
228	the Zayetkwin area (Figure 2b). They are medium- to coarse-grained with granoblastic
229	and polygonal texture, and are characterized by the assemblage of Cal + Dol + Chu +
230	Fo + Spl + Amp + Phl + Gr (Figure 3) and Cal + Dol + Fo + Spl + Phl, respectively
231	(Figure 4a and b). Calcite is coarse-grained (0.2–1 mm in size) and usually occurs as a
232	matrix phase (Figures 3a-c and e, and 4a and b) and as inclusions in clinohumite
233	(Figure 3a), spinel (Figures 3d and 4a), and forsterite (Figure 4b). In KST03, calcite
234	also occurs as a retrograde product replacing dolomite (Figure 3c). The calcite grains
235	included by clinohumite and other phases exhibit exsolution particles of dolomite. The
236	matrix calcite grains in ZYK01 show a dolomite exsolution texture (Figure 4a and b) as
237	do calcite inclusions. However, in KST03, the matrix (Figure 3a, c) and retrograde
238	(Figure 3c) calcite grains, do not show such an exsolution texture, except for a narrow
239	marginal zone (100–200 $\mu$ m in width) around some matrix grains (Figure 3e). Dolomite
240	grains usually occur as exsolution products in calcite (Figures 3d and 4a, d) and isolated
241	grains in the matrix (Figures 3c and 4a-c), which might be partly the result of
242	coalescence of exsolved dolomite. In KST03, dolomite grains additionally occur as
243	reaction products between calcite and clinohumite (Figure 3a and b). The dolomite
244	grains of reaction products are texturally divided into those that occur as isolated zones
245	(Figure 3a and b) and as symplectitic aggregates with tremolite (Figure 3a, b, and f).
246	The dolomite-tremolite symplectites rarely contain diopside (Figure 3a and e), in which
247	diopside is always surrounded by tremolite (Figure 3e).
248	Spinel grains occur as prismatic porphyroblasts (Figures 3c and 4a: up to 3 mm in
249	diameter) and inclusions (200 $\mu$ m in width) in clinohumite (Figure 3b). In ZYK01,

250 forsterite grains occur as stable phases and tend to form irregular- or granular-shaped 251 grains (0.3–3 mm in diameter) (Figure 4b), and partly pseudomorphed or altered to 252 serpentine (Figure 4a and b). In contrast, only small granular-shaped forsterite 253 inclusions (50–500  $\mu$ m) were observed in the clinohumite porphyroblasts in KST03 254 sample (Figure 3a and b). Granular- or irregular-shaped clinohumite grains (2-3 mm in 255 size) occur as a major phase in KST03 and are always rimmed by a zone which consists 256 of dolomites and symplectitic aggregates of tremolite and dolomite (Figure 3a, b, and f). 257 This intergrowth zone borders clinohumite with calcite, texturally indicating a reaction 258 product of clinohumite and calcite. Most clinohumite grains include small and granular 259 forsterite grains indicating that these two phases were in equilibrium during the 260 prograde stage (Figure 3a and b). Although tremolite commonly forms symplectite with 261 dolomite around clinohumite crystals (Figure 3f), primary pargasite/edenite is also 262 observed around spinel grains in association with phlogopite and chlorite in KST03 263 (Figure 3c). Phlogopite also occurs as a matrix phase in ZYK01 (Figure 4a). Chlorite 264 flakes occur between spinel and clinohumite/dolomite grains (Figure 3b) and as rim 265 phase around pargasite/edenite (Figure 3c).

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#### 267 **3.2** | Forsterite-free marble

268 The forsterite-free marble samples (OZ01 and ACT01) were collected from calc-silicate

and diopside marble unit in the Onzon (96°05'32" E, 22°48'50" N) and Aung Tharyar

270 (96°00'19" E, 22°53'08" N) areas, respectively (Figure 2b). The main mineral

- assemblages of these samples, OZ01 and ACT01, are Cal + Dol + Spl + Phl (Figure 4c)
- and Cal + Di + Phl + Prg (Figure 4e), respectively. In OZ01, calcite grains, which
- 273 exhibit an exsolution texture with dolomite, are the major matrix phase and also occur

274 as inclusions in spinel (Figure 4c and d). Although relatively coarse dolomite grains (up 275 to 700–800  $\mu$ m) were observed in the matrix of OZ01, they were always surrounded by 276 calcite or occurred as an interstitial phase between calcite and silicate phases (Figure 4c and d). Therefore, the coarse-grained dolomite in the matrix might have been the result 277 278 of coalescence of exsolved dolomite; there are no isolated primary dolomite grains in 279 OZ01. Calcite grains of both inclusion and matrix phases in ACT01 do not exhibit a 280 significant dolomite exsolution texture, although some parts of the matrix grains contain 281 small amounts of exsolved dolomite (Figure 4f). 282 Prismatic spinel grains (up to 0.6 mm in size) are observed in OZ01 and commonly 283 include calcite grains with dolomite exsolution particles (Figure 4c). Large flakes of 284 phlogopite are dominant in the matrix in both samples (Figure 4c and e), and also 285 observed as aggregates associated with spinel in OZ01 and with diopside in ACT01. 286 Diopside grains occur as granular- or anhedral-shaped grains of 0.3-1 mm in size in 287 ACT01. They commonly form aggregates with calcite, phlogopite, and pargasite, and 288 some grains include isolated calcite grains (Figure 4e). In ACT01, secondary tremolite 289 partially replaces diopside around the margins. Chlorite occurs as a secondary phase 290 replacing some fine-grained spinel grains in OZ01.

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#### 292 **3.3** | Calc-silicate rock

293 Calc-silicate rock samples were collected from the calc-silicate and diopside marble unit

294 in the Onzon (OZ03: 96°05'37" E, 22°48'48" N) and Wabyudaung (WBY01:

295 96°06'34" E, 22°52'07" N) areas, and the paragneiss unit in the Thabeikkyin

296 (95°58'59" E, 22°53'18" N) area (Figure 2b). They are fine to medium-grained and

show granoblastic textures. The main mineral assemblage of these samples is Scp + Di

298 + Cal + Qz + Kfs + Ttn (Figure 5a). Scapolite is a major primary phase in all 299 calc-silicate rock samples and occurs as subhedral to anhedral grains of 0.1-0.7 mm in 300 size. Some scapolite grains in OZ03 show optical zoning consisting of a primary core 301 and a thin meionite rim (Figure 5a). The primary scapolite grains in WBY01and KB01 302 were retrogressively replaced by plagioclase with minor amounts of calcite and quartz 303 (Figure 5b) and clinozoisite (Figure 5c), respectively. Coarse plagioclase grains, which 304 are partly replaced by aggregates of muscovite and quartz, occur as inclusions in calcite 305 of WBY01 (Figure 5d). An aggregate of plagioclase with scapolite and/or K-feldspar 306 was also included in the calcites of WBY01, and no distinct reaction relationship was 307 observed between them (Figure 5b). These textural characteristics suggest that these 308 plagioclase grains were primary phase and in equilibrium with scapolite. The matrix 309 diopside grains are rounded granular matrix minerals and range from 0.4 - 1.7 mm in 310 size. Those in OZ03 show subhedral porphyroblasts and enclose scapolite, calcite, 311 quartz, and titanite. Although most of the diopside grains in OZ03 are partly replaced by 312 tremolite, those in WBY01 and KB01 show no replacement or pseudomorph texture. In 313 OZ03 and WBY01, quartz is the dominant phase and coexists with scapolite, diopside 314 and calcite (Figure 5a), whereas the quartz modal composition of KB01 is less than 1 315 vol%. Clinozoisite occurs only in KB01, which is texturally differentiated into two 316 domains: one is dominated by scapolite, diopside, titanite, and small amounts of quartz 317 and calcite and the other is characterized by almost monomineralic clinozoisite (> 3 mm 318 in diameter) with scapolite and calcite relics (Figure 5c). Titanite grains occur as 319 rounded and subhedral crystals in the matrix of all of the studied samples (Figure 5a). 320 They also occur as inclusions in the scapolite and diopside/clinozoisite of OZ03 and 321 KB01, respectively.

323 4 I MINERAL CHEMISTRY

324 The constituent minerals in the marble and calc-silicate rock were analyzed using the 325 JEOL JXA-8800R (WDS + EDS) electron-probe microanalyzer (EPMA) at the 326 Department of Earth and Planetary Sciences of Nagoya University. The accelerating 327 voltage and specimen current for the quantitative analyses were maintained at 15 kV 328 and 12 nA on the Faraday cup, respectively. Beam diameters of 10 and 5  $\mu$ m were used 329 for calcite and mica/feldspar analysis, respectively, and diameters of  $2-3 \mu m$  were used 330 for the analyses of all other phases. Well-characterized natural and synthetic phases, 331 including synthetic F-phlogopite (F = 8.7 wt%) for F and natural Cl-rich hastingsite (Cl 332 = 3.27 wt%: Suwa, Enami & Horiuchi, 1987) for Cl, were employed as standards. 333 Matrix corrections were performed using the  $\alpha$ -factor table of Kato (2005). The 334 representative chemical analyses of the major constituent minerals of the marble and 335 calc-silicate rock samples are listed in Tables 2 and 3, respectively. The Fe<sup>3+</sup>/Fe<sup>2+</sup> value 336 for amphibole was estimated considering an average of the minimum and maximum 337 values proposed by Leake et al. (1997) and that for spinel was estimated assuming 338 stoichiometry and a charge balance. The total iron content was assumed to be in the 339 form of Fe<sub>2</sub>O<sub>3</sub> for clinozoisite and titanite and FeO for the other phases. 340

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## **341 4.1 | Clinohumite**

Clinohumite grains from the Mogok marble are compositionally homogeneous and have an  $M_{Ti}/Si$  value of 2.22 ± 0.02 (Figure 6), where  $M_{Ti}$  denotes the total octahedral cations including Ti (Gaspar, 1992; Jones, Ribbe & Gibbs, 1969). This compositional range is consistent with those of titanian clinohumite from the carbonatites of the Jacupiranga

Complex of Brazil (2.22 ± 0.03: Gaspar, 1992) and F-rich clinohumite from the Ambasamudram marble of Southern India (2.22 ± 0.01 for low  $M_{Tr}/Si$  group data: Satish-Kumar & Niimi, 1998). The humite group minerals consist of laminations of olivine and brucite layers, and their compositions are characterized by the number "*n*", which indicates the ratio of these two layers [*n*(M, Fe)<sub>2</sub>SiO<sub>4</sub>•(Mg, Fe)(F, OH)<sub>2</sub>] (Jones *et al.*, 1969). From their stoichiometry, the relationship of  $M_{Tr}/Si$  with *n* is

- $\frac{352}{Si} \qquad \qquad \frac{M_{Ti}}{Si} = \frac{(2n+1)}{n}$
- 353

354 Most of the  $M_{Ti}/Si$  values from the Mogok samples fall below the expected value of 2.25

355 (n = 4) that would be the minimum for the humite group minerals, indicating the

356 presence of superstructures with n > 4 in the analyzed clinohumite (e.g., Gaspar, 1992).

357 The spots with lower  $M_{Ti}/Si$  values correspond to clinohumite phases where the stacking

358 sequence is mainly made up of n = 4 and 6 in different proportions for each

359 superstructure (e.g., White & Hyde, 1982). The  $TiO_2$  and FeO contents are less than

360 0.15 wt% and 0.36 wt%, respectively, and  $X_{Mg}$  [= Mg/(Mg + Fe)] = 1.00. The F and Cl

361 contents are 3.57–4.14 wt% [ $X_F = F/2$  for O = 17  $\approx F/(F + Cl + OH) = 0.62 \pm 0.03$ ] and

- less than 0.02 wt%, respectively.
- 363

#### 364 4.2 | Phlogopite

365 Phlogopite grains have near Mg-endmember compositions ( $X_{Mg} = 0.96-1.00$ ) and are

rich in F (1.4–3.6 wt%). The average  $X_F$  values for KST03, ZYK01, OZ01, and ACT01

367 are  $0.77 \pm 0.06$ ,  $0.38 \pm 0.03$ ,  $0.44 \pm 0.04$ , and  $0.50 \pm 0.03$ , respectively (Figure 7a).

- 368 Chlorine contents are typically less than 0.05 wt% in all samples. The average TiO<sub>2</sub>
- 369 content varies from  $0.16 \pm 0.02$  wt% in ZYK01 to  $0.81 \pm 0.05$  wt% in OZ01. Although

370 the CaO content is less than 0.1 wt% in all samples, BaO and Na<sub>2</sub>O contents reach 7.9 371 wt% [0.22 atomic per formula unit (apfu) for O = 11] and 1.36 wt% (0.19 apfu) in 372 KST03 (Figure 7b). The phlogopite grains in ZYK01 also contain up to 1.1 wt% (0.16 373 apfu) Na<sub>2</sub>O. Barium incorporation into Ti-poor phlogopite is generally controlled by the 374 following substitutions:  $Ba \square K_{-2}$  and  $BaAlK_{-}Si_{-1}$ , where  $\square$  indicates a vacant site (e.g., 375 Fleet, 2003). The (Ba + Ca) - (K + Na) diagram suggests that Ba-substitution of 376 phlogopite in KST03 is promoted by the combination of these two types of substitutions 377 (Figure 7c). (Ba + Ca) > (3-Si) compositions (for O = 11) in some Ba-rich grains 378 (Figure 7d) are consistent with this interpretation. However, the compositional trend of 379 some Ba-rich grains with (Ba + Ca) < (3-Si) cannot be explained by these Ba-bearing 380 substitutions alone. Considering the compositional trend subparallel to the Si-axis, the 381 compositional varieties of the Ba-rich phlogopite of KST03 were mainly controlled by 382 combinations of three types of substitutions, including the tschermakite substitution 383  $(Al_2Mg_{-1}Si_{-1}).$ 

384

385 4.3 | Forsterite and spinel

386 Forsterite grains have endmember compositions of  $X_{Mg} = 0.99 - 1.00$ , and their NiO,

387 MnO, and CaO contents are below the detection limit of about 0.03 (CaO)–0.04 (NiO

 $388 \quad \text{ and } MnO) \text{ wt\% } (2\sigma \text{ level}).$ 

389 Spinel grains also have near endmember compositions of  $X_{Mg} = 0.90-1.00$  and  $X_{Al}$  [=

390  $Al/(Al + Fe^{3+} + Cr + V)] = 0.98-1.00$ . The V<sub>2</sub>O<sub>3</sub> content reaches 1.3 wt% in KST03, and

391 the ZnO content is up to 2.2 wt% in OZ01. These components, however, are usually less

than 0.1 wt% and 0.5 wt% in other samples, respectively.

### 394 4.4 | Scapolite, plagioclase, and K-feldspar

395 Primary scapolite grains are relatively homogeneous and exhibit a solid solution mainly 396 between meionite (Ca<sub>4</sub>Al<sub>6</sub>Si<sub>6</sub>O<sub>24</sub>CO<sub>3</sub>) and mizzonite (NaCa<sub>3</sub>Al<sub>5</sub>Si<sub>7</sub>O<sub>24</sub>CO<sub>3</sub>), with 397 equivalent anorthite (EqAn) = 65-72 and  $X_{Cl} = 0.05-0.07$  in OZ03; EqAn = 73-77 and 398  $X_{Cl} = 0.01-0.02$  in WBY01; and EqAn = 69-73 and  $X_{Cl} = 0.08-0.11$  in KB01, where  $EqAn = (Al-3)/3 \times 100$  and  $X_{Cl} = Cl/2$  for total cations =  $16 \approx Cl/(Cl + CO_3)$  (Figure 8). 399 400 The primary core of scapolite in OZ03 was retrogressively rimmed by a marialite 401 (Na<sub>4</sub>Al<sub>3</sub>Si<sub>9</sub>O<sub>24</sub>Cl)-rich mantle with EqAn = 33-51 and  $X_{Cl} = 0.42-0.78$  (Figure 5a). In 402 KB01, scapolite was strongly replaced by clinozoisite (Figure 5c) and has a relatively 403 marialite-richer composition (EqAn = 40-65 and  $X_{Cl} = 0.18-0.51$ ) than the primary 404 phase.

405 Texturally, primary plagioclase grains (Figure 5b and d) are homogeneous and have 406 mostly near endmember compositions of  $An_{90-96}$  (Figure 8). In contrast, plagioclase 407 grains, which retrogressively replaced scapolite (Figure 5b), have a slightly sodic 408 composition with  $An_{75-85}$ . The orthoclase contents of K-feldspar vary from  $Or_{87-93}$  in 409 OZ03 to  $Or_{94-96}$  in KB01. The BaO and CaO contents are less than 0.4 wt% and 0.2 wt%,

410 respectively.

411

#### 412 **4.5** | **Amphibole**

413 Amphibole grains in the marble (KST03, ZYK01, and ACT01) are Mg-endmember

414 varieties with  $X_{Mg} = 0.92 - 1.00$ . They are texturally grouped into primary and secondary

415 phases. The primary phases are pargasite/edenite with Si values down to 6.00 apfu (O =

416 23) and Al and <sup>[A]</sup>(Na + K) values up to 2.90 apfu and 0.97 apfu, respectively, where

417	<sup>[A]</sup> (Na + K) indicates the alkaline contents at the largest A-site (Figure 9a). In contrast,
418	secondary phases, which replace primary amphibole or are products of the retrogressive
419	reaction of clinohumite and calcite (Figure 3f), are tremolite with Si values mostly
420	greater than 7.48 apfu and Al and $^{[A]}(Na + K)$ values less than 0.94 apfu and 0.41 apfu,
421	respectively. The amphibole grains in the marble are characteristically rich in F with $X_F$
422	= 0.27-0.54 (1.2-2.5  wt%) for the primary phase and $0.11-0.32 (0.51-1.54  wt%)$ for
423	the secondary phase (Figure 9b). Chlorine contents are less than 0.09 wt% for all grains.
424	Amphibole grains in the calc-silicate rock (OZ03), which replace diopside, are F-poor
425	tremolite and have slightly lower $X_{Mg}$ values (0.75–0.82) than those in the marble: Si =
426	7.50–7.87 apfu, Al = 0.20–0.77 apfu, <sup>[A]</sup> (Na + K) = 0.01–0.12 apfu, $X_F < 0.02$ (< 0.07
427	wt%), and Cl < 0.02 wt%.
428	
429	4.6   Calcite and dolomite
429 430	<ul><li><b>4.6</b>   Calcite and dolomite</li><li>Calcite grains in marble are simple solid solutions between calcite and dolomite with</li></ul>
430	Calcite grains in marble are simple solid solutions between calcite and dolomite with
430 431	Calcite grains in marble are simple solid solutions between calcite and dolomite with FeO and MnO contents of mostly less than 0.2 wt% (0.3 mol% of siderite) and 0.05
430 431 432	Calcite grains in marble are simple solid solutions between calcite and dolomite with FeO and MnO contents of mostly less than 0.2 wt% (0.3 mol% of siderite) and 0.05 wt% (0.1 mol% of rhodochrosite), respectively. The matrix calcite grains with
<ul><li>430</li><li>431</li><li>432</li><li>433</li></ul>	Calcite grains in marble are simple solid solutions between calcite and dolomite with FeO and MnO contents of mostly less than 0.2 wt% (0.3 mol% of siderite) and 0.05 wt% (0.1 mol% of rhodochrosite), respectively. The matrix calcite grains with exsolution textures in ZYK01 and OZ01 are usually divided into core and mantle parts,
<ul> <li>430</li> <li>431</li> <li>432</li> <li>433</li> <li>434</li> </ul>	Calcite grains in marble are simple solid solutions between calcite and dolomite with FeO and MnO contents of mostly less than 0.2 wt% (0.3 mol% of siderite) and 0.05 wt% (0.1 mol% of rhodochrosite), respectively. The matrix calcite grains with exsolution textures in ZYK01 and OZ01 are usually divided into core and mantle parts, which are rich and poor in dolomite particles, respectively (Figure 4a, c, and d). The
<ul> <li>430</li> <li>431</li> <li>432</li> <li>433</li> <li>434</li> <li>435</li> </ul>	Calcite grains in marble are simple solid solutions between calcite and dolomite with FeO and MnO contents of mostly less than 0.2 wt% (0.3 mol% of siderite) and 0.05 wt% (0.1 mol% of rhodochrosite), respectively. The matrix calcite grains with exsolution textures in ZYK01 and OZ01 are usually divided into core and mantle parts, which are rich and poor in dolomite particles, respectively (Figure 4a, c, and d). The mantle is usually poorer in magnesite content than the core (Figure 4d). Although the
<ul> <li>430</li> <li>431</li> <li>432</li> <li>433</li> <li>434</li> <li>435</li> <li>436</li> </ul>	Calcite grains in marble are simple solid solutions between calcite and dolomite with FeO and MnO contents of mostly less than 0.2 wt% (0.3 mol% of siderite) and 0.05 wt% (0.1 mol% of rhodochrosite), respectively. The matrix calcite grains with exsolution textures in ZYK01 and OZ01 are usually divided into core and mantle parts, which are rich and poor in dolomite particles, respectively (Figure 4a, c, and d). The mantle is usually poorer in magnesite content than the core (Figure 4d). Although the exsolved parts of the matrix and inclusion are slightly heterogeneous, the variation of
<ul> <li>430</li> <li>431</li> <li>432</li> <li>433</li> <li>434</li> <li>435</li> <li>436</li> <li>437</li> </ul>	Calcite grains in marble are simple solid solutions between calcite and dolomite with FeO and MnO contents of mostly less than 0.2 wt% (0.3 mol% of siderite) and 0.05 wt% (0.1 mol% of rhodochrosite), respectively. The matrix calcite grains with exsolution textures in ZYK01 and OZ01 are usually divided into core and mantle parts, which are rich and poor in dolomite particles, respectively (Figure 4a, c, and d). The mantle is usually poorer in magnesite content than the core (Figure 4d). Although the exsolved parts of the matrix and inclusion are slightly heterogeneous, the variation of magnesite content is 1–2 mol% in most cases (e.g., Figure 3d). Calcite grains, which
<ul> <li>430</li> <li>431</li> <li>432</li> <li>433</li> <li>434</li> <li>435</li> <li>436</li> <li>437</li> <li>438</li> </ul>	Calcite grains in marble are simple solid solutions between calcite and dolomite with FeO and MnO contents of mostly less than 0.2 wt% (0.3 mol% of siderite) and 0.05 wt% (0.1 mol% of rhodochrosite), respectively. The matrix calcite grains with exsolution textures in ZYK01 and OZ01 are usually divided into core and mantle parts, which are rich and poor in dolomite particles, respectively (Figure 4a, c, and d). The mantle is usually poorer in magnesite content than the core (Figure 4d). Although the exsolved parts of the matrix and inclusion are slightly heterogeneous, the variation of magnesite content is 1–2 mol% in most cases (e.g., Figure 3d). Calcite grains, which contain exsolved dolomite particles, have relatively low magnesite components of less

441	and c). Isolated calcite grains without dolomite spherules in the matrix of KST03
442	(Figure 3a, c, and e) have relatively low magnesite contents: up to 7.0 mol%. The
443	matrix calcite grains in ACT01 mostly occur as isolated phases and show little
444	exsolution texture. In the case of locally exsolved calcite in ACT01, the host calcite
445	around the dolomite spherules exhibit locally high calcite content (Figure 4f). However,
446	calcite in points 100–200 $\mu$ m away from the dolomite has lower calcite composition,
447	and shows continuous variation of up to $9.8 \text{ mol}\%$ of the magnesite component (Figure
448	10d). ACT01 contains no primary dolomite in the matrix or the inclusion phase,
449	indicating that the matrix calcite was not saturated in the magnesite component during
450	its formation. The wide compositional ranges and rare development of exsolution
451	textures of the matrix calcite grains in ACT01 might be largely attributed to the
452	undersaturated magnesite environment during the prograde metamorphic stage. The
453	calcite grains included in the diopside of ACT01 do not show exsolution textures and
454	have lower magnesite content (up to 2.7 mol%) than the matrix grains. Texturally
455	retrograde calcite in KST03, which replaces matrix dolomite (Figure 3c), has similar
456	magnesite content (up to 6.4 mol%) to the isolated matrix calcite (Figure 10a).
457	Calcite grains in the calc-silicate rock have near endmember compositions, and MgO,
458	FeO, and MnO are less than 0.7 wt% (1.5 mol% of magnesite), 1.1 wt% (1.5 mol% of
459	siderite), and 0.2 wt% (0.2 mol% of rhodochrosite), respectively.
460	Dolomite grains in marble, except those in ACT01, are in simple solid solution, and
461	the FeO and MnO contents are less than 0.3 wt% and 0.05 wt%, respectively. In ACT01,
462	the FeO content reaches 2.5 wt% (3.4 mol% of siderite) and MnO was not detected.
463	Calcite content varies from 52 mol% of ZYK01 to 57 mol% of ACT01 on average, and

464 does not show distinct differences between exsolved spherules and the isolated phase in465 the matrix.

466 Homogeneous calcite compositions prior to the unmixing phenomenon in marble 467 were estimated by reintegration based on: firstly, the average chemical compositions of 468 calcite and dolomite; secondly, their volume ratio estimated using digital back-scattered 469 electron (BSE) images and the image analysis software 'ImageJ 1.51g' (Rasband, W.S., 470 ImageJ, U. S. National Institutes of Health, Bethesda, Maryland, 471 USA, http://imagej.nih.gov/ij/, 1997-2012) (Schneider, Rasband & Eliceiri, 2012); and 472 thirdly, densities of calcite and dolomite of 2.71 and 2.87 g/cm<sup>3</sup>, respectively, which 473 were calculated using lattice constants reported by Paquette and Reeder (1990). The 474 estimated volume ratios ( $V_{Dol}/V_{Cal}$ ) of the inclusion phases are 0.18–0.55, and those of 475 the matrix phases vary from 0.22–0.24 in OZ01 to <0.01 in ACT01. 476 In KST03, the primary calcite grains included in clinohumite have distinctly more 477 magnesite-rich compositions (20.4 –21.2 mol%) than those in spinel (12.5–13.0 mol%). 478 Conversely, the reintegrated compositions of exsolved calcite grains in the other two 479 samples are relatively constant within each sample and do not show any relationship to 480 their modes of occurrence: ZYK01, 13.3-14.1 mol% for matrix grains, 13.0 and 14.3 481 mol% for inclusions in spinel, 14.0 and 16.4 mol% for inclusions in forsterite; OZ01, 482 12.0 and 12.7 mol% for matrix grains and 13.2–14.5 mol% for inclusions in spinel. 483 Although matrix calcite grains in ACT01 locally show exsolution texture, their 484 reintegrated compositions were not estimated because of the very low volume contents 485 of the exsolved dolomite particles ( $V_{Dol}/V_{Cal} < 0.01$ ). However, it is possible that the 486 primary compositions of the calcite grains might have been similar to those of the

487 magnesite-rich part of the isolated grains free of dolomite spherules.

489	4.7   Diopside
490	Primary diopside grains in marble (ACT01) have near endmember composition with
491	$X_{Mg} = 0.90-0.98$ and TiO <sub>2</sub> and Na <sub>2</sub> O contents less than 0.4 wt% and 0.3 wt%,
492	respectively. The $Al_2O_3$ contents vary from 0.5 to 5.2 wt% and tend to be higher in the
493	inclusions in phlogopite and other silicates than the isolated phase in the matrix. The
494	secondary diopside grains in marble (KST03) have purer endmember compositions than
495	the primary grains in ACT01: $X_{Mg} = 0.99 - 1.00$ , TiO <sub>2</sub> < 0.1 wt%, Na <sub>2</sub> O < 0.1 wt%, and
496	Al <sub>2</sub> O <sub>3</sub> < 0.4 wt%. The primary diopside grains in the calc-silicate rock have lower $X_{Mg}$
497	values (0.63–0.86: FeO = 4.6–11.7 wt%) and $Al_2O_3$ content (0.6–1.9 wt%) than those in
498	the marble. TiO <sub>2</sub> and Na <sub>2</sub> O contents are less than 0.3 wt% and 0.2 wt%, respectively.
499	
500	4.8   Other phases
501	The $X_{Fe}$ [= Fe <sup>3+</sup> /(Fe <sup>3+</sup> + Al)] values of clinozoisite in a calc-silicate rock (KB01) are
502	0.06–0.18, and the MnO contents are less than 0.15 wt%. Titanite grains are relatively
503	homogeneous and do not have distinct differences in compositions between inclusions

504 and the matrix phases (KB01):  $Al_2O_3 = 2.4-2.8$  wt%,  $Fe_2O_3 = 0.7$  wt% and F = 1000

505 0.55-0.83 wt% for the inclusion phase and Al<sub>2</sub>O<sub>3</sub> = 2.3-2.9 wt%, Fe<sub>2</sub>O<sub>3</sub> = 0.5-0.8 wt%

and F = 0.39-0.65 wt% for the matrix phase. In the three calc-silicate rock samples, the

507  $Al_2O_3$ ,  $Fe_2O_3$ , and F contents are 2.3–4.1 wt%, 0.2–0.8 wt%, and 0.7–1.1 wt%,

508 respectively. Chlorine content is below the detection limit of 0.03 wt%.

509

## 510 **5** I CALCITE-DOLOMITE SOLVUS GEOTHERMOMETER

511	Calcite-dolomite solvus thermometry was both experimentally and thermodynamically
512	characterized by Anovitz and Essene (1987), Goldsmith and Newton (1969) and Powell,
513	Condliffe and Condliffe (1984). As the calcite-dolomite solvus geothermometer is
514	almost independent of pressure, it is known as a reliable thermometer for marble of
515	amphibolite facies (e.g., Letargo, Lamb & Park, 1995; Rathmell, Streepey, Essene &
516	van der Pluijm, 1999; Suzuki, 1977; Wada & Suzuki, 1983). However, Mg-rich calcite
517	grains in granulite facies rocks typically show exsolution textures, suggesting
518	re-equilibrium during the retrograde stage (e.g., Mizuochi, Satish-Kumar, Motoyoshi &
519	Michibayashi, 2010).
520	Most calcite grains in the Mogok marble show exsolution textures and give
521	re-equilibrated temperature conditions during retrograde stage of 350-610 °C for
522	KST03, 370–540 °C for ZYK01 and 360–550 °C for OZ01, employing the calibration
523	proposed by Anovitz and Essene (1987). However, some Mg-richer spots yield higher
524	temperatures of 610–680 °C in ZYK01 (Figure 10b) and 600–660 °C in OZ01 (Figure
525	10c). The isolated calcite grains without exsolution textures in KST03 record low
526	temperatures of 370–600 °C, similar to the exsolved inclusions (Figure 10a). The calcite
527	replacing dolomite also shows temperatures of 400–580 °C. The isolated grains in
528	ACT01, in contrast, show a continuous variation from 355 to 670 °C (Figure 10d), and
529	inclusion calcite grains show lower temperature conditions of less than 410 °C.
530	The primary compositions of calcite grains estimated by reintegration of the exsolved
531	calcite and dolomite aggregate yield higher and slightly scattered temperature ranges of
532	720-880 °C. In KST03, calcite grains included in clinohumite show distinctly higher
533	temperature conditions (870–880 °C) than those in spinel (730–740 °C) (Figure 10a). In
534	contrast, no distinct difference in temperature conditions owing to the difference of

535	modes of occurrence of calcite was observed in ZYK01 and OZ01. In ZYK01, the
536	matrix calcite and the inclusion calcites in forsterite and spinel record temperatures of
537	745–760 °C, 735–760 °C, and 755–800 °C, respectively. In OZ01, calcite grains in the
538	matrix and in spinel show temperature conditions of 720-730 °C and 740-765 °C,
539	respectively. In KST03 and ZYK01, isolated dolomite grains occur as a matrix phase
540	and coexist with the exsolved calcite and dolomite aggregate. Therefore, the original
541	Mg-richer calcite grains estimated for these samples have probably been in equilibrium
542	with dolomite, which might represent the peak temperature conditions.
543	In OZ01, relatively coarse dolomite grains (up to 700–800 $\mu$ m) occur in the matrix.
544	However, they are probably coalescent phases of exsolution spherules as described in
545	Section 3.2 (Figure 4c and d), and thus, the original Mg-richer calcite grains in OZ01
546	probably did not coexist with dolomite at the peak metamorphic stage. Therefore,
547	temperature estimates of OZ01 (720-765 °C) based on the reintegrated Mg-rich calcite
548	compositions probably define the lower limit of temperature conditions of the peak
549	metamorphic stage. ACT01 contains no isolated dolomite in the matrix, and the matrix
550	calcite grains showing temperatures up to 670 °C also define the minimum conditions
551	of peak metamorphism. The minimum temperatures estimated by OZ01 and ACT01 are
552	slightly lower than those estimated for the reintegrated compositions of KST03 and
553	ZYK01 (730–880 °C).
554	Ye Kyaw Thu et al. (2016, 2017) estimated the equilibrium temperature conditions of
555	paragneiss from the Onzon and Thabeikkyin areas as 780-860 °C at 0.60–0.84 GPa
556	based on conventional geothermobarometers employing partial assemblages of Grt + Bt
557	+ Pl + Crd + Spl + Sill + Qz and empirical Ti-in-biotite and Zr-in-rutile
558	geothermometers. Yonemura et al. (2013) also reported equilibrium conditions of

559 0.65–0.87 GPa/800–950 °C for garnet-orthopyroxene granulite samples in the Mogok

560 area. The temperature conditions of metacarbonate rocks estimated by the

561 calcite-dolomite geothermometer are consistent with those of the surrounding

562 lithologies.

563

#### 564 6 | DISCUSSION

565 6.1 | Peak equilibria and retrograde reactions

566 The relationships of *P*-*T*-fluid composition were calculated using

567 THERMOCALC software ver. 3.33 (Powell & Holland, 1988) and an internally

568 consistent thermodynamic data set (Holland & Powell, 1998; updated June 2009).

569 Chemical compositions of minerals employed for the estimations of the metamorphic

570 conditions are listed in Table 4. The activities of the solid solution phases are obtained

571 from the following sources: scapolite, plagioclase, clinozoisite, diopside, and F-free

572 tremolite (AX\_2 program;

573 http://www.esc.cam.ac.uk/research/research-groups/research-projects/tim-hollands-soft

574 <u>ware-pages/ax</u>), clinohumite (Rice, 1980), and F and Mg-rich tremolite in KST03

575  $[(1-X_F)^2]]$ . Unit activity was used for other phases unless otherwise noted. The chemical

576 formulae of minerals employed for reactions discussed on the following pages are after

577 Holland and Powell (1998). The pressure condition at peak granulite facies stage was

578 assumed to be 0.8 GPa, considering equilibrium conditions estimated for the paragneiss

579 surrounding the marble and calc-silicate rock (Ye Kyaw Thu *et al.*, 2016, 2017).

580

581 6.1.1 | CaO-MgO-SiO<sub>2</sub>-H<sub>2</sub>O-CO<sub>2</sub> system of marble

582 In KST03, F-bearing clinohumite occurs as a main constituent mineral and includes 583 forsterite and calcite (Figure 3a and b). The assemblage of these phases is 584 characteristically stable under high-temperature and F-bearing conditions (Rice, 1980; 585 Satish-Kumar *et al.*, 2001). Figure 11 presents  $T-X_{CO2}$  diagrams showing the stability of 586 these and related phases in the system CaO-MgO-SiO<sub>2</sub>-CO<sub>2</sub>-H<sub>2</sub>O with excess calcite. 587 Ba-rich phlogopite, pargasite/edenite, and spinel, which are major Al-bearing phases in 588 KST03, have not been considered. 589 The sample KST03 contains dolomite as an isolated matrix phase, forsterite as an 590 inclusion phase in clinohumite, and calcite as both the isolated matrix and inclusion 591 phases. These textural relationships imply that forsterite, calcite and dolomite coexisted

in equilibrium before and during the formation of clinohumite, and clinohumite formedby the following reaction:

594 
$$4Fo + Dol + H_2O = Chu + Cal + CO_2$$
. R1

This reaction would have resulted in a systematic increase of  $X_{CO2}$  along the univariant curve R1 with increasing temperature in a closed system. However, the limited occurrence of forsterite only as inclusions in clinohumite (Figure 3a and b) implies that forsterite was not in equilibrium with dolomite and the inclusion + host assemblage of Chu + Fo + Cal was stable at the peak metamorphic stage. The stability field of this assemblage is defined by the combination of reaction R1 and the following reaction:

601 
$$14Fo + Cal + 3H_2O = 3Chu + Di + CO_2$$
. R2

The matrix assemblage of clinohumite + calcite + dolomite was also stable under the estimated peak metamorphic condition (Fig. 11a). These facts imply that the  $X_{CO2}$  value of fluid was firstly internally buffered by the reaction R1, and thus, probably increased with increasing temperature at the beginning of clinohumite formation, but subsequently

606 decreased, which destabilized the Fo + Dol assemblage, at the peak metamorphic stage,

although the mechanism of the change in fluid composition remains to be explained.

608 The lower temperature and  $X_{CO2}$  limits of the Chu + Fo + Cal assemblage are estimated

as about 810 °C and 0.60 for KST03 (invariant point [Tr] in Figure 11a).

610 A zone consisting of dolomite and a symplectitic aggregate of tremolite and dolomite

611 constantly develops between the clinohumite and matrix calcite (Figure 3a and b),

612 although retrograde diopside rimmed by tremolite rarely joins the aggregate (Figure 3a

and e). The common occurrence of diopside-free tremolite and dolomite symplectite

614 suggests that the following reaction mainly controlled the breakdown of the Chu + Cal

615 assemblage during the retrograde stage:

616 
$$2Chu + 15Cal + 11CO_2 \rightarrow Tr + 13Dol + H_2O$$
R3

617 However, this reaction is metastable for a wide range of  $X_{CO2}$  conditions, at least > 0.001, 618 under the higher pressure of > 0.7 GPa, thus an alternative reaction R4, controls the 619 stability of the Chu + Cal assemblage under the assumed wide  $X_{CO2}$  conditions (Figure 620 11a):

621 
$$Chu + 9Cal + 5CO_2 \rightarrow 2Di + 7Dol + H_2O$$
 R4

The rare occurrence of diopside replaced by tremolite in the aggregate of tremolite and dolomite (Figure 3e) probably implies that the reaction R4 progressed locally before proceeding to the reaction R3. The reaction R3 is stable under lower pressure of < 0.7 GPa and  $X_{co2}$  conditions, and its upper stability is defined by the invariant point [Fo], which shifts from 0.002/490 °C at 0.65 GPa, through 0.084/620 °C at 0.5 GPa, to 0.25/640 °C at 0.4 GPa with decreasing pressure (Figure 11b). This fact suggests that the reaction between clinohumite and calcite shown in Figure 3a, b, and f have 629 progressed during the retrograde stage under distinctly lower  $X_{CO2}$  conditions than 630 during the peak metamorphic stage.

631 ZYK01 is composed of forsterite, calcite, and dolomite without diopside. The 632 stability of this assemblage is defined by the combination of reaction R1 and the 633 following reaction.

634

635

$$Di + 3Dol = 2Fo + 4 Cal + 2CO_2$$
 R5

The Fo + Cal + Dol assemblage is stable at higher  $X_{CO2}$  conditions than the Chu + Fo + 636 Cal assemblage observed in KST03 for the same compositional conditions (cf., Figure

637 11a). Its stability field, however, shifts towards lower temperature and  $X_{CO2}$  conditions

638 with increasing activity, decreasing  $X_F$ , of clinohumite. For example,  $X_{CO2}$  and

639 temperature conditions of the invariant point [Tr] for  $X_F(Chu) = 0.5$  are 0.004 and

640 530 °C, respectively, at 0.8 GPa. Therefore, these two assemblages may have shared the

641 same temperature and  $X_{CO2}$  conditions, if the F content of the metamorphic fluid of

642 ZYK01 were relatively lower than that of KST03. The lower limit of the  $X_{CO2}$  condition

643 for the stability of Fo + Cal + Dol assemblage cannot be estimated because of the

644 absence of clinohumite in ZYK01. However, this assemblage in ZYK01 does not

645 conflict with the high temperature and  $X_{CO2}$  estimates of the peak metamorphic stage for

646 KST03, which was collected from the Kyetsaung Taung marble unit, as was ZYK01

647 (Figure 2b). In ZYK01, the coexisting forsterite, calcite, and dolomite do not show any

648 reaction such as R5 during the retrograde stage. This might imply that the  $X_F$  value of

649 metamorphic fluid of ZYK01 was lower than that of KST03 even during the retrograde

650 stage, and thus: the stability field of the clinohumite-bearing assemblage reduced and

651 that of the Fo + Cal + Dol assemblage relatively expanded towards lower  $X_{CO2}$  and

652 temperature conditions; and the  $X_{CO2}$  value was relatively low during the retrograde

656 under the high temperature and  $X_{CO2}$  conditions during granulite facies stage deduced 657 from the mineral assemblages of KST03 and ZYK01 (Figure 11a). 658 659 6.1.2 | CaO-MgO-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub>-H<sub>2</sub>O-CO<sub>2</sub> system of calc-silicate rock 660 The characteristic mineral assemblages of the calc-silicate rock at the peak granulite 661 facies stage are Scp + Pl + Di + Qz + Cal for WBY01 and Scp + Di + Qz + Cal for 662 OZ03 and KB01. Figure 12 shows  $T-X_{C02}$  diagram that demonstrate the stability of 663 scapolite and related phases of WBY01 and OZ03 in the CaO-Al2O3-SiO2-CO2-H2O 664 system with excess quartz and calcite. Figure 13 shows  $T-X_{CO2}$  diagrams of scapolite 665 and related phases of KB01 in the CaO-MgO-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub>-CO<sub>2</sub>-H<sub>2</sub>O system during the 666 peak granulite facies and the retrograde stage. 667 The peak metamorphic conditions of the scapolite-bearing calc-silicate rock are 668 defined by the following reactions: 669 3An + Cal = MeiR6 670  $Mei + 3Qz + 5Cal = 3Grs + 6CO_2$ **R**7 671  $Qz + Cal = Wo + CO_2$ **R**8 672 Temperature and X<sub>CO2</sub> conditions of the invariant points defined by these reactions 673 calculated employing unit activity for grossular ([Wo] and [Wo, Czo] in Figures 12 and 674 13, respectively) are 795 °C/0.22, 780 °C/0.17, and 790 °C/0.19 for WBY01, OZ03, and 675 KB01, respectively. The invariant point [Wo] shifts towards higher  $X_{CO2}$  conditions

stage, for example, the Fo + Cal + Dol assemblage is stable under  $X_{CO2} > 0.05$  at 0.5

CaO-MgO-SiO<sub>2</sub> system of OZ01 and Di + Cal assemblage of ACT01 are also stable

GPa/600 °C for  $X_F$  (*Chu*) = 0.5. The silicate mineral-free assemblage in the

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676 under constant temperature with decreasing grossular composition of garnet in the

677 system (Figure 12), and thus, the isobaric diagram calculated assuming unit activity of 678 grossular defines the minimum  $X_{CO2}$  condition. Therefore, temperature and  $X_{CO2}$ 679 conditions at the peak metamorphic stage of WBY01, which contains a Scp + Pl + Qz +680 Cal assemblage, were estimated as 795 °C and > 0.22, respectively, at P = 0.8 GPa 681 (Figure 12). Conversely, OZ03 and KB01 contain no prograde plagioclase, and thus, the 682 Scp + Qz + Cal assemblage defines the minimum temperature and  $X_{CO2}$  conditions of 683 780 °C and 0.17 for OZ03 (Figure 12) and 790 °C and 0.19 for KB01 (Figure 13a), 684 respectively, at P = 0.8 GPa. These estimates of the lower limit conditions are 685 consistent with the estimates by the Scp + Pl + Qz + Cal equilibrium for WBY01. 686 In KB01, clinozoisite occurs forming aggregate with quartz and calcite, and thus, the 687 retrograde trajectory might have passed the stability field of the Czo + Qz + Cal688 assemblage defined by the following reactions R9 and R10: 689  $2Czo + 3Qz + 5Cal = 3Grs + H_2O + 5CO_2$ R9 690  $2Czo + CO_2 = 3An + Cal + H_2O$ R10 691 This stability field is located at lower temperature and expands towards lower X<sub>CO2</sub> 692 condition than that of the Scp + Qz + Cal assemblage under the same pressure, and 693 shifts towards lower temperature condition with decreasing pressure (Figure 13a). In 694 Figure 13a, the reactions R9 was calculated assuming pure grossular composition, and

the univariant line R9 and invariant point [Mei, Wo] shift toward lower temperature and
higher X<sub>CO2</sub> conditions with decreasing activity of grossular. Therefore, the stability

697 field of the Czo + Qz + Cal assemblage in Figure 13a defined by the combination of

698 reactions R9 and R10 shows the maximum temperature and minimum  $X_{CO2}$  value of the

699 stability of clinozoisite in the calc-silicate rock (KB01). Additionally, diopside occurs

700 coexisting with the Czo + Qz + Cal assemblage without tremolite, suggesting that the

701 temperature and  $X_{CO2}$  conditions of the Czo + Di + Qz + Cal assemblage's stability were 702 defined by the combination of R9, R10 and the following reaction: 703  $Tr + 2Qz + 3Cal = 5Di + H_2O + 3CO_2$ R11 704 Consequently, the temperature and  $X_{CO2}$  of the main retrograde stage based on the 705 stability of the Czo + Di + Qz + Cal assemblage are estimated at < 620 °C and < 0.155 706 at P = 0.5 GPa, respectively (Figure 13a). 707 In KB0, clinozoisite also occurs replacing scapolite (Figure 5c), implying the 708 possible following retrograde reaction: 709 Mei + H<sub>2</sub>O  $\rightarrow$  Czo + CO<sub>2</sub> R12 710 This reaction is stable only in quartz-absent metacarbonate rock, because, in the 711 quartz-excess system, the stability of scapolite is defined by reactions R6 and R7, and 712 that of clinozoisite is controlled by alternative reactions R9 and R10 as discussed above 713 (Figure 13a). Figure 13b shows stability of the reaction R12 in the quartz-free system. 714 An invariant point [Grs, Wo], including reaction R12, is located at high temperature/low 715  $X_{CO2}$  conditions of 750 °C/0.059 for P = 0.8 GPa and almost isothermally decreases with 716 decreasing pressure down to 745°C/0.009 at P = 0.4 GPa (Figure 13b). These 717 metamorphic conditions imply that wollastonite should have been stable instead of Qz + 718 Cal assemblage (R8) if the reaction R12 has stably progressed during the retrograde 719 stage. However, the wide occurrence of Qz + Cal assemblage and absence of 720 wollastonite in metacarbonate rocks throughout the studied Onzon-Thabeikkyin region 721 are inconsistent with the temperature and  $X_{CO2}$  conditions supposed by the reaction R12. 722 Consequently, the replacement of scapolite by clinozoisite in KB01 might have not 723 progressed in stable under the high temperature and low  $X_{CO2}$  conditions in the

SiO<sub>2</sub>-undersaturated system, and probably suggests metastable reaction in the stability field of Qz + Cal assemblage.

In a calc-silicate rock (WBY01), scapolite grains were replaced by plagioclase and
calcite with small amounts of quartz (Figure 5b), which follows reaction R6. If the
quartz grains were a breakdown product of scapolite, the following reaction might be
more suitable, as discussed by Motoyoshi et al. (1991):

730 
$$Scp \rightarrow Pl + Qz + Cal$$
 R13

They considered that the precipitation of quartz is due to a small excess of  $SiO_2$  in the scapolite over the ideal composition, or, alternatively,  $SiO_2$  was released together with Na<sub>2</sub>O from the marialite component.

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#### 735 **6.2** | Evolution of metamorphic fluids

Figure 14 summarizes the *P*-*T* conditions of the peak granulite facies and subsequent low-amphibolite facies stage, and compares the compositions of metamorphic fluids of the marble and the calc-silicate rock of the Mogok metamorphic belt between the two stages of recrystallizations. Low  $X_{CO2}$  fluid composition of paragneiss surrounding the metacarbonate rocks, which is inferred based on their carbonate minerals-absent assemblages (Maw Maw Win *et al.*, 2016; Ye Kyaw Thu *et al.*, 2016, 2017), was also shown for comparison.

743

## 744 6.2.1 | Peak granulite facies stage

745 The mineral assemblages of the metacarbonate rocks in the Onzon–Thabeikkyin region

recorded the minimum temperature conditions of 780–810 °C at 0.8 GPa for the peak

747 metamorphic stage. The calcite-dolomite solvus geothermometer also suggested a

748	minimum temperature of 720–765 °C and possible equilibrium temperature up to
749	870-880 °C. These estimates are consistent with those for peak granulite facies
750	metamorphism recorded in the paragneiss of the Onzon-Thabeikkyin region (0.60-0.84
751	GPa/780-860 °C) (Ye Kyaw Thu et al., 2016, 2017). These data suggest that the
752	metacarbonate rocks and the surrounding paragneiss shared similar P-T metamorphic
753	conditions with the granulite facies. However, fluid compositions at the peak
754	metamorphic stage seem to have been different among the lithologies. The $X_{CO2}$ values
755	of the metamorphic fluid were significantly higher ( $> 0.60$ in the marble and $>$
756	0.17–0.22 in the calc-silicate rock) than that of a nearly pure $H_2O$ -fluid inferred for the
757	paragneiss. Although it is not conclusive that the $X_{CO2}$ values were different between the
758	marble and calc-silicate rock at peak metamorphic stage, there were significant
759	discontinuities in the metamorphic fluid compositions between the metacarbonate rocks
760	and paragneiss.

#### 762 6.2.2 | Retrograde stage

The recrystallization textures of granulite facies assemblages (Figures 3a, b, e, and f and 5c) during the retrograde stage imply that the  $X_{CO2}$  values of the metamorphic fluid decreased with decreasing metamorphic *P*-*T* conditions. For the discussions of temperature and  $X_{CO2}$  conditions during retrograde recrystallization, the pressure was assumed to be 0.5 GPa because the paragneiss in the Onzon–Thabeikkyin region were extensively recrystallized at P = 0.40-0.55 GPa with fluid infiltration (Ye Kyaw Thu *et al.*, 2016, 2017).

The retrograde reaction between clinohumite and calcite (reaction R3) observed in

771 the marble (KST03) probably progressed at  $X_{CO2}$  and temperature conditions lower than

772 0.084 and 620 °C at P = 0.5 GPa, respectively (Figure 11b). The stability of Fo + Cal + 773 Dol assemblage without Di throughout the peak and retrograde metamorphic stages in 774 ZYK01 also suggest decrease of  $X_{CO2}$  condition with decreasing temperature as 775 discussed in section 6.1.1. The coexisting clinozoisite, diopside, quartz, and calcite of 776 KB01 estimate the  $X_{CO2}$  and temperature conditions as < 620 °C and < 0.155 at P = 0.5777 GPa, respectively (Figure 13a), also suggesting lower  $X_{CO2}$  condition at retrograde stage 778 than that at the peak granulite facies stage. 779 Consequently, the metacarbonate rocks studied herein recorded a distinct gap of fluid 780 composition between the peak and retrograde stages (Figure 14). Ye Kyaw Thu et al. 781 (2016, 2017) suggested that the Mogok paragneiss in the Onzon area have recrystallized 782 to varying degree with extensive infiltration of H<sub>2</sub>O-rich fluids during exhumation 783 (0.40-0.55 GPa/620-640 °C). A similar extensive hydration stage was also recorded in 784 the paragneiss from the Mandalay region (0.3-0.5 GPa/600-680 °C) by Maw Maw Win 785 et al. (2016) and Enami, Nagaya and Maw Maw Win (2017). Although there is no 786 conclusive evidence suggesting that the retrograde recrystallizations of the 787 metacarbonate rocks developed under conditions similar to those of the paragneiss, 788 there is no inconsistency in the P-T estimations of the retrograde stage between the two 789 lithologies. Episodic infiltration of H<sub>2</sub>O-rich fluid during exhumation probably enabled 790 the retrograde reactions of the metacarbonate rocks with low X<sub>CO2</sub>-fluid phases under 791 low-amphibolite facies conditions. 792

793

### 6.3 | Contrasts in fluid compositions

794 The metamorphic fluid of the Mogok metacarbonate rocks had distinctly higher  $X_{CO2}$ 795 composition than the H<sub>2</sub>O-dominant fluid of the paragneiss at the peak metamorphic

796	stage. There might have also been $X_{CO2}$ contrast between the marble and calc-silicate
797	rock. Yui, Zaw and Wu (2008) analysed $\delta^{I_3}C - \delta^{I_8}O$ compositions of ruby-bearing and
798	related marble from the Mogok metamorphic belt, and concluded that the formation of
799	ruby might have resulted from CO2-rich fluid-rock interaction, while spinel- and
800	forsterite-bearing marble was genetically recrystallized under low $X_{CO2}$ environment.
801	Some major constituent minerals in the marble and calc-silicate rock are richer in F and
802	Cl, respectively. The mafic minerals in the marble have nearly Mg-endmember
803	compositions ( $X_{Mg} = 0.92 - 1.00$ ) and those in the calc-silicate rock are more enriched in
804	FeO ( $X_{Mg} = 0.63-0.86$ for diopside). Therefore, the distinct differences of halogen
805	compositions were certainly controlled by the Fe-F and Mg-Cl avoidance principles.
806	However, the halogen compositions of minerals also reflected the characteristics of the
807	fluid compositions and probably indicate the contrast of the halogen compositions of
808	metamorphic fluids during the peak granulite facies stage between these metacarbonate
809	lithologies. These features suggest that the metamorphic fluid compositions were
810	internally buffered and the effective migration of metamorphic fluids, which causes
811	extensive change in the fluid compositions beyond the major lithologic boundaries, did
812	not occur during the peak granulite facies stage throughout the Mogok metamorphic
813	belt.
814	Similar local fluid buffering during peak metamorphism was well documented in
815	metacarbonate rocks from many granulite facies terranes (e.g., Kaneko et al., 2005;
816	Satish-Kumar et al., 1996; Satish-Kumar et al., 2001). These studies also concluded that
817	$X_{CO2}$ values of the metamorphic fluids were probably maintained as high as 0.5–0.8 even
818	during the low-granulite and amphibolite facies stages of the retrograde stage. In

819 contrast, in the case of the Mogok metamorphic belt, the far-ranging infiltration of

820	$H_2O\text{-rich}$ fluids during exhumation and hydration stage probably decreased the $X_{\text{CO2}}$
821	value of the metacarbonate rocks and extensively homogenized the metamorphic fluid
822	compositions throughout the metacarbonate rocks and the surrounding paragneiss. Maw
823	Maw Win et al. (2016) discussed the relationship between the chemical zoning of
824	monazite and CHIME ages of each zone in a Mogok paragneiss, and considered that the
825	monazite ages were reset to a varying degree at ca. 24 Ma due to fluid infiltration. In the
826	Mogok metamorphic belt, extensive acidic magmatic intrusions were reported during
827	late Oligocene-early Miocene by zircon U-Pb ages (e.g., Barleyet al., 2003; Mitchell,
828	Chung, Thura Oo, Lin and Hung, 2012; Searle et al., 2007). Although the origin of the
829	inferred H <sub>2</sub> O-dominant fluid during the exhumation has not been conclusively
830	established, released fluid from the intruded and solidified melt might have contributed
831	to the modification of the fluid composition. Alternatively, the fluid might have been
832	released from a slab dehydrated during active subduction.

## 834 **7 I SUMMARY**

835 The following points summarize this study of the metacarbonate rocks from the Mogok836 metamorphic belt:

1 The metacarbonate rocks from the middle segment of the Mogok metamorphic belt
studied are grouped into the dolomite-bearing and quartz-free marble and the
dolomite-free and quartz-bearing calc-silicate rock. The halogen contents of minerals
show distinct contrast between the marble and calc-silicate rock. Hydrous phases in
marble are characteristically rich in F: clinohumite up to 4.1 wt%, phlogopite up to
3.6 wt%, prograde amphibole up to 2.5 wt%, and retrograde amphibole up to 1.5
wt%. In contrast, calc-silicate rock is not composed of a F-bearing phase and

844		scapolite grains characteristically contain Cl up to $0.5 \text{ wt\%}$ in the prograde phase and
845		3.1 wt% in the retrograde phase. Some phlogopite grains in marble have unusually
846		high BaO and Na <sub>2</sub> O contents of up to 7.9 wt% and 1.4 wt%, respectively.
847	2	Thermodynamic analyses of reaction relations of minerals show that the
848		metacarbonate rocks were equilibrated under high temperature conditions of >
849		780–810 °C at 0.8 GPa, which are equivalent to the surrounding paragneiss.
850		However, $X_{CO2}$ values of metamorphic fluids of the metacarbonate rocks during the
851		peak granulite facies stage were distinctly higher than those of the surrounding
852		paragneiss: $X_{CO2} > 0.6$ for marble and $> 0.17-0.22$ for calc-silicate rock. The high
853		$X_{CO2}$ compositions during the prograde stage decreased to $X_{CO2} < 0.08$ for marble and
854		< 0.155 for calc-silicate rock at around 0.5 GPa/600 °C during retrograde stage.
855	3	The differences in halogen compositions of constituent minerals and the estimated
856		$X_{CO2}$ values of metamorphic fluids between the marble and calc-silicate rock imply
857		that compositions of metamorphic fluids during the granulite facies stage were
858		internally buffered. Migration of these fluids, which effectively caused
859		inhomogeneity in the composition, did not progress beyond the lithological
860		boundaries among these metacarbonate rocks and the surrounding paragneiss.
861		Subsequently, the fluid compositions were extensively homogenized and became low
862		$X_{CO2}$ during the retrograde stage by infiltration of H <sub>2</sub> O-rich fluids. The hydrous fluids
863		probably originated from one or both of the following sources: (1) fluids released
864		from acidic melts that intruded at a later stage of metamorphism; and/or (2)
865		dehydration of a slab undergoing subduction.
866		

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- 1115 Captions for Figures
- FIGURE 1. Geological map of Myanmar and the adjacent regions showing major suture
  zones, faults, and terrane boundaries [modified from Figure 1 of Mitchell *et al.*(2007)].
- FIGURE 2. Geological maps of (a) the middle segment of the Mogok metamorphic belt
  and the adjacent regions [part of Figure 2 of Mitchell *et al.* (2012)] and (b)
- detailed geological map of the sampling areas with sample localities
- 1122 [unpublished geological data by Myint Lwin Thein *et al.* (1990)].
- 1123 FIGURE 3. Back-scattered electron (BSE) and X-ray mapping images showing textural
- 1124 characteristics of major constituent minerals of a marble (KST03) from the
- 1125 Mogok metamorphic belt. (a) and (b) Clinohumite and its inclusions and
- 1126 reaction relationship between clinohumite and calcite, (c) aggregates of
- 1127 phlogopite, pargasite, and secondary chlorite in matrix, (d) calcite grain with
- dolomite spherules included in a spinel, and (e) tremolite, diopside, and dolomite
- aggregate and (f) symplectitic aggregate of tremolite and dolomite between
- 1130 clinohumite and calcite. Numbers in (d) indicate mol% of magnesite component
- 1131 in calcite. Abbreviation for mineral: Tr-symp, symplectitic aggregate of
- tremolite and dolomite.
- FIGURE 4. Back-scattered electron (BSE) and X-ray mapping images showing textural
  characteristics of major constituent minerals of marble from the Mogok
- 1135 metamorphic belt. (a) and (b) modes of occurrences of major phases of ZYK01,
- 1136 (c) calcite grains occurring as inclusions in spinel and the matrix phase of OZ01,
- 1137 (d) variations of Ca content of the matrix calcite showing exsolution textures of
- 1138 OZ01, (e) mode of occurrence of major phases of ACT01, and (f) variations of

1139	Ca content of the matrix calcite, which locally contains exsolved dolomite of
1140	ACT01. Numbers in (f) indicate mol% of magnesite component in calcite.
1141	FIGURE 5. Back-scattered electron (BSE) and X-ray mapping images showing textural
1142	characteristics of major constituent minerals of calc-silicate rock from the
1143	Mogok metamorphic belt. (a) Scapolite with sodic rim coexisting with diopside,
1144	calcite, K-feldspar, quartz, and titanite (OZ03), (b) Scapolite partly replaced by
1145	plagioclase (S-Pl), calcite, and small amounts of quartz (WBY01), (c) scapolite
1146	extensively replaced by clinozoisite (KB01), and (d) anorthite-rich plagioclase
1147	partly replaced by an aggregate of muscovite and quartz (WBY01). "Sodic Scp"
1148	indicates sodic scapolite rim around calcic scapolite. "P-Pl" and "S-Pl" imply
1149	primary (An <sub>90-96</sub> ) and secondary (An <sub>75-85</sub> ) plagioclase grains, respectively.
1150	Numbers indicate $EqAn$ (a and c) and $An$ (b and d) contents of representative
1151	analytical points.
1152	FIGURE 6. Cumulative diagrams of (a) $M_{Ti}/Si$ value and (b) F (wt %) of clinohumite in
1153	a marble from the Mogok metamorphic belt (KST03). $M_{Ti}$ and "n" indicate total
1154	octahedral cations including Ti (Gaspar, 1992; Jones et al., 1969) and the ratio of
1155	the olivine layer to the brucite layer $[nMg_2SiO_4 \bullet Mg(F, OH)_2]$ , respectively.
1156	FIGURE 7. Compositional range of phlogopite in marble from the Mogok metamorphic
1157	belt in (a) F (wt%) cumulative number, (b) K–Na–(Ba + Ca), (c) (Ba + Ca)–(K +
1158	Na), and (d) Si–(Ba + Ca) diagrams. $\Box$ denotes vacant-site.
1159	FIGURE 8. Compositional range of scapolite and plagioclase in calc-silicate-rock from
1160	the Mogok metamorphic belt in EqAn [= (Al-3)/3 × 100] $-X_{Cl}$ [= Cl/2 for total
1161	cations = $16 \approx Cl/(Cl + CO_3)$ ] diagram. Lines bracket the stoichiometric range of
1162	scapolite grains proposed by Evans, Shaw and Haughton (1969).

FIGURE 9. Compositional range of amphibole in marble and calc-silicate rock from the Mogok metamorphic belt in (a)  $Si^{[A]}(Na + K)$  and (b)  $Si^{-X_F} = F/2 \approx F/(F + Cl + OH)$ ] diagrams.

- FIGURE 10. Cumulative diagrams of magnesite mol% of calcite in marble from the
  Mogok metamorphic belt. Data showing magnesite content less than 2 mol% are
  omitted. Each integrated composition was estimated using dataset of a whole
  inclusion or core part of an exsolved matrix grain. Temperature conditions were
- estimated using a geothermometer proposed by Anovitz and Essene (1987).
- 1171 FIGURE 11. Isobaric  $T-X_{CO2}$  diagrams in the calcite-excess system for the
- clinohumite-bearing marble of KST03 from the Mogok metamorphic belt at
- 1173 pressures of (a) 0.8 GPa and (b) 0.5 GPa. Fixed activities of clinohumite at
- 1174 temperatures of 800 °C and 600 °C were employed for calculations of the
- diagrams (a) and (b), respectively. The pressure dependence of the upper
- 1176 stability limit of  $X_{CO2}$  and temperature conditions of the reaction R3 Tr + 13Dol
- $H_2O = 2Chu + 15Cal + 11CO_2$  is shown in (b) for the pressure range of
- 1178 0.4–0.65 GPa. Labels R1–R5 correspond to reactions discussed in the text. H<sub>2</sub>O
- and CO<sub>2</sub> components of reactions are omitted for convenience. Minerals other
- than quartz, which are in equilibrium with calcite, are shown in CaO–MgO–SiO<sub>2</sub>
  diagrams.
- 1182FIGURE 12. Isobaric  $T-X_{CO2}$  diagram in the quartz and calcite-excess system for the1183scapolite-bearing calc-silicate rock of WBY01 and OZ03 from the Mogok1184metamorphic belt showing stability relationship of scapolite and related phases1185at P = 0.8 GPa. Fixed activities of meionite and anorthite at temperatures of1186800 °C, and that of grossular calculated as  $(X_{Grs})^3$ , assuming ideal solid solution
  - 51

1187 (where  $X_{Grs}$  implies molecular proportion of grossular), were employed for

calculation of the diagrams. H<sub>2</sub>O and CO<sub>2</sub> components of reactions are omitted
for convenience. Labels R6–R8 correspond to those of the reactions discussed in
the text.

1191	FIGURE 13. Isobaric $T-X_{CO2}$ diagrams for the scapolite-bearing calc-silicate rock of
1192	KB01 from the Mogok metamorphic belt showing stability relationships of
1193	scapolite and related phases (a) in the quartz and calcite-excess system at $P = 0.8$
1194	GPa and (b) in the quartz-free and calcite-excess system under various pressure
1195	conditions between 0.4 and 0.8 GPa. Univariant lines of $Wo = Qz + Cal$ at 0.5
1196	and 0.8 GPa are superimposed on the diagram (b) to compare the stability
1197	conditions of Qz + Cal and Czo. Fixed activities of phases at temperatures of
1198	800 °C and 750 °C were employed for calculations of the diagrams (a) and (b),
1199	respectively. H <sub>2</sub> O and CO <sub>2</sub> components of reactions are omitted for convenience.
1200	Labels R6–R12 correspond to those of the reactions discussed in the text.
1201	Broken lines with arrow in (a) and (b) indicate shift trend of invariant points
1202	[Mei, Wo] and [Grs, Wo], respectively.
1203	FIGURE 14. Schematic diagram showing evolution of fluid compositions of marble,
1204	calc-silicate rock, and paragneiss from the middle segment of the Mogok
1205	metamorphic belt in Myanmar. The stabilities of aluminum silicates are from
1206	Pattison (2001). References for P-T estimations of paragneiss are Y16, Ye Kyaw
1207	Thu et al. (2016); Y17, Ye Kyaw Thu et al. (2017).
1208	
1209	

1210 Caption for Tables

1211	Table 1. Mineral assemblages in marble and calc-silicate rock from the
1212	Onzon-Thabeikkyin region of the Mogok metamorphic belt, Myanmar.
1213	+, prograde phase in matrix; i, inclusion phase in primary phase; ei, exsolved
1214	phase in calcite inclusion; em, exsolved phase in matrix calcite; r, retrograde
1215	phase; ce, coalescence phase of exsolved dolomite.
1216	Note: 1) In the matrix calcite, exsolution texture is observed only as narrow
1217	marginal zone (100–200 $\mu$ m in width) in some grains, 2) Calcite inclusion in
1218	diopside do not show exsolution texture.
1219	Table 2. Representative chemical compositions of major constituent minerals of
1220	marble from the Onzon-Thabeikkyin region of the Mogok metamorphic belt,
1221	Myanmar.
1222	* Total iron as FeO.
1223	** Calculated values.
1224	Abbreviations for texture: Pgp, prograde phase; Rgp, retrograde phase; Symp,
1225	retrograde symplectitic phase.
1226	Table 3. Representative chemical compositions of major constituent minerals of
1227	calc-silicate rock from the Onzon-Thabeikkyin region of the Mogok
1228	metamorphic belt, Myanmar.
1229	* Total iron as FeO.
1230	** Total iron as $Fe_2O_3$ .
1231	† Total cations as 16.
1232	§ Calculated value (see text).
1233	Abbreviations for texture: Pgp, prograde phase; Rgp, retrograde phase.

1234	Table 4. Chemical compositions of minerals employed for the calculations of $T-X_{C02}$
1235	diagrams.
1236	* Anorthite content was estimated assuming that An/EqAn = 1.2 considering
1237	the pair of prograde scapolite and plagioclase in WBY01.
1238	** $X_{Mg}$ value was assumed to be same with that of diopside.
1239	Abbreviations for texture: Pgp, prograde phase; Rgp, retrograde phase.

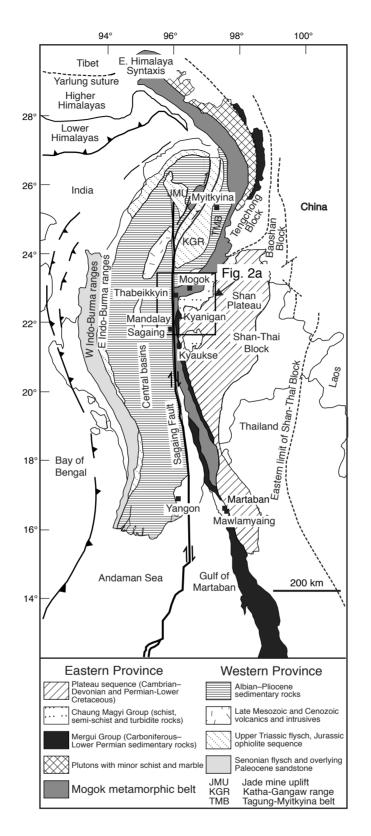


FIGURE 1. Geological map of Myanmar and the adjacent regions showing major suture zones, faults, and terrane boundaries [modified from Figure 1 of Mitchell et al. (2007)].

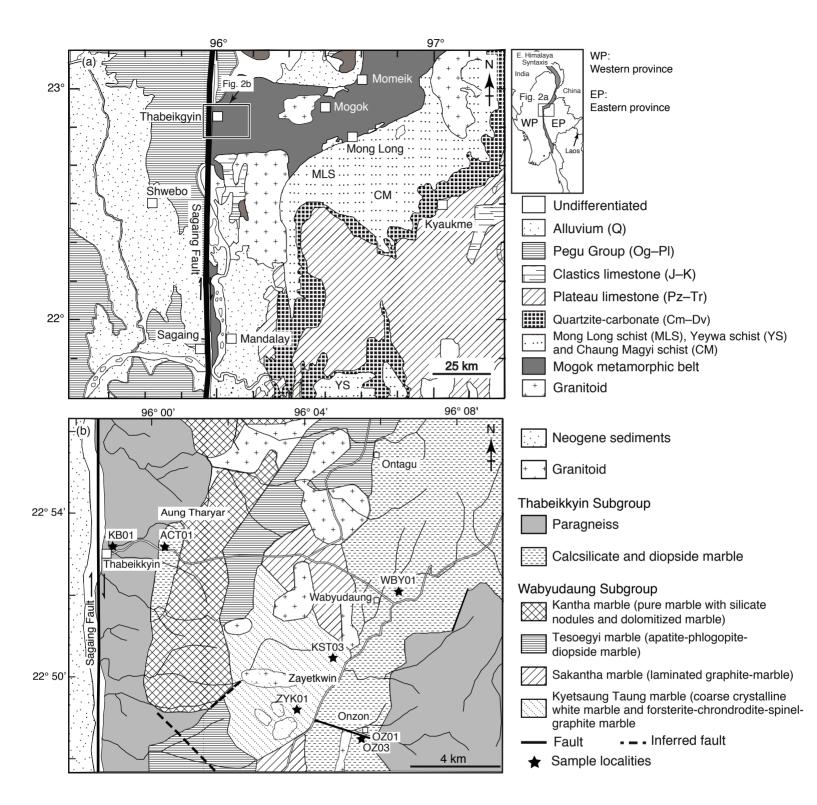


FIGURE 2. Geological maps of (a) the middle segment of the Mogok metamorphic belt and the adjacent regions [part of Figure 2 of Mitchell et al. (2012)] and (b) detailed geological map of the sampling areas with sample localities [unpublished geological data by Myint Lwin Thein et al. (1990)].

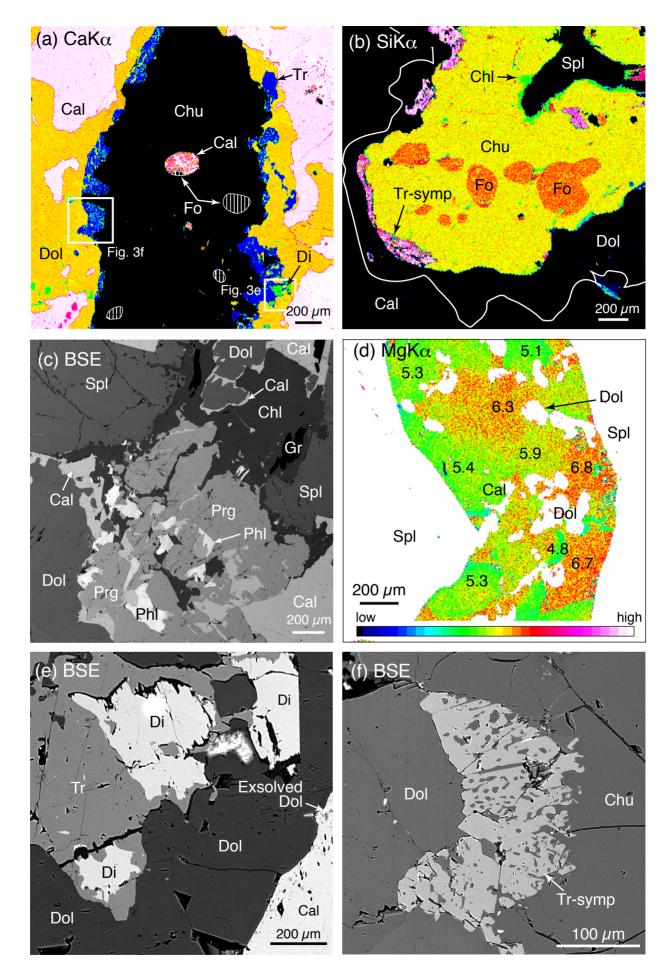


FIGURE 3. Back-scattered electron (BSE) and X-ray mapping images showing textural characteristics of major constituent minerals of a marble (KST03) from the Mogok metamorphic belt. (a) and (b) Clinohumite and its inclusions and reaction relationship between clinohumite and calcite, (c) aggregates of phlogopite, pargasite, and secondary chlorite in matrix, (d) calcite grain with dolomite spherules included in a spinel, and (e) tremolite, diopside, and dolomite aggregate and (f) symplectitic aggregate of tremolite and dolomite between clinohumite and calcite. Numbers in (d) indicate mol% of magnesite component in calcite. Abbreviation for mineral: Tr-symp, symplectitic aggregate of tremolite and dolomite.

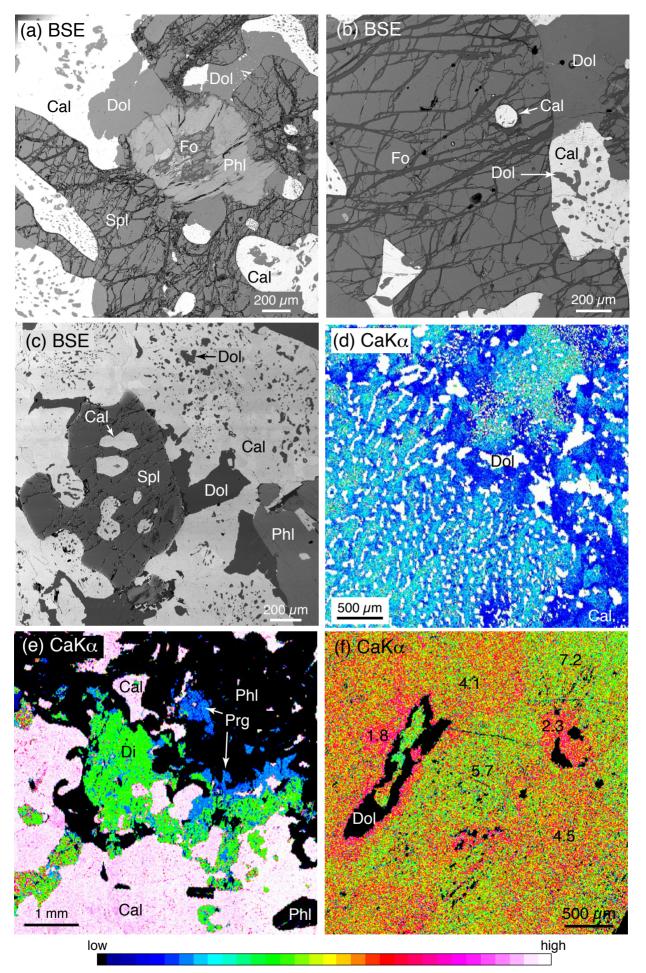


FIGURE 4. Back-scattered electron (BSE) and X-ray mapping images showing textural characteristics of major constituent minerals of marble from the Mogok metamorphic belt. (a) and (b) modes of occurrences of major phases of ZYK01, (c) calcite grains occurring as inclusions in spinel and the matrix phase of OZ01, (d) variations of Ca content of the matrix calcite showing exsolution textures of OZ01, (e) mode of occurrence of major phases of ACT01, and (f) variations of Ca content of the matrix calcite, which locally contains exsolved dolomite of ACT01. Numbers in (f) indicate mol% of magnesite component in calcite.

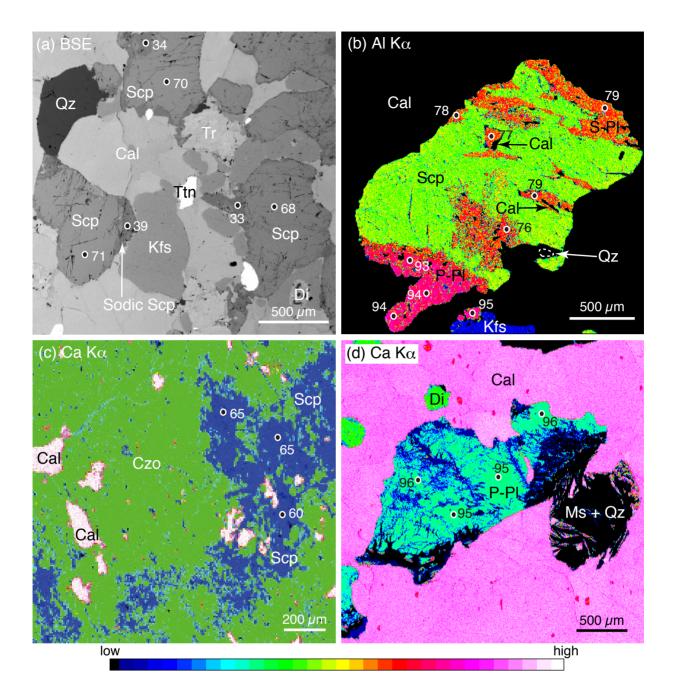


FIGURE 5. Back-scattered electron (BSE) and X-ray mapping images showing textural characteristics of major constituent minerals of calc-silicate rock from the Mogok metamorphic belt. (a) Scapolite with sodic rim coexisting with diopside, calcite, K-feldspar, quartz, and titanite (OZ03), (b) Scapolite partly replaced by plagioclase (S-PI), calcite, and small amounts of quartz (WBY01), (c) scapolite extensively replaced by clinozoisite (KB01), and (d) anorthite-rich plagioclase partly replaced by an aggregate of muscovite and quartz (WBY01). "Sodic Scp" indicates sodic scapolite rim around calcic scapolite. "P-PI" and "S-PI" imply primary (An90–96) and secondary (An75–85) plagioclase grains, respectively. Numbers indicate EqAn (a and c) and An (b and d) contents of representative analytical points.

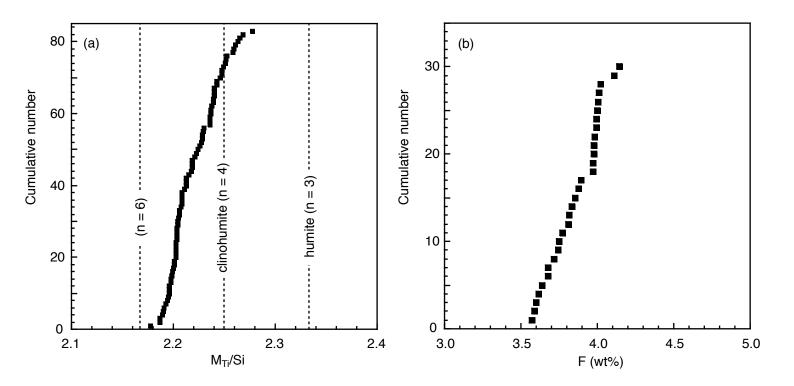


FIGURE 6. Cumulative diagrams of (a) MTi/Si value and (b) F (wt %) of clinohumite in a marble from the Mogok metamorphic belt (KST03). MTi and "n" indicate total octahedral cations including Ti (Gaspar, 1992; Jones et al., 1969) and the ratio of the olivine layer to the brucite layer  $[nMg_2SiO_4\cdot Mg(F, OH)_2]$ , respectively.

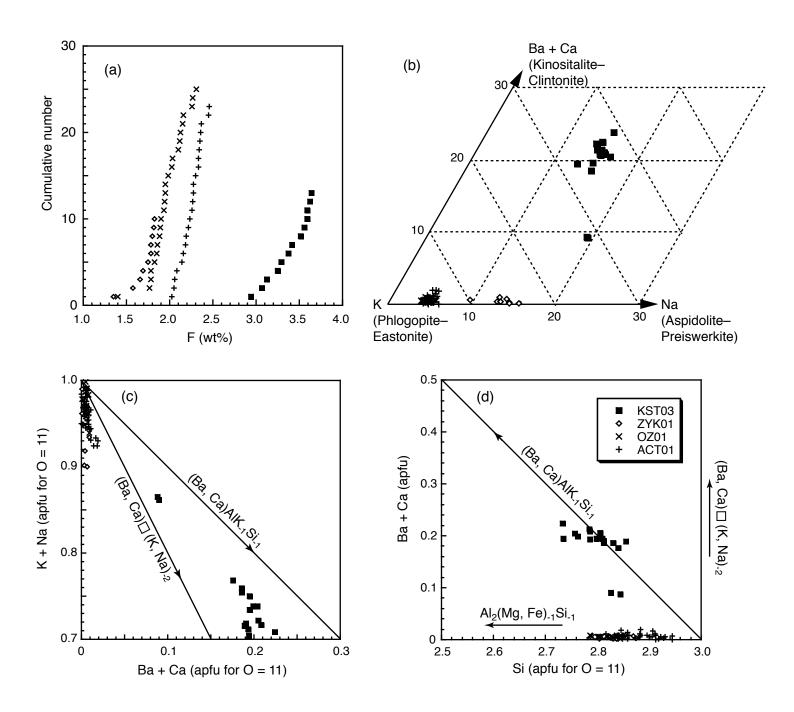


FIGURE 7. Compositional range of phlogopite in marbles from the Mogok metamorphic belt in (a) F (wt%) cumulative number, (b) K–Na–(Ba + Ca), (c) (Ba + Ca)–(K + Na), and (d) Si–(Ba + Ca) diagrams.  $\Box$  denotes vacant-site.

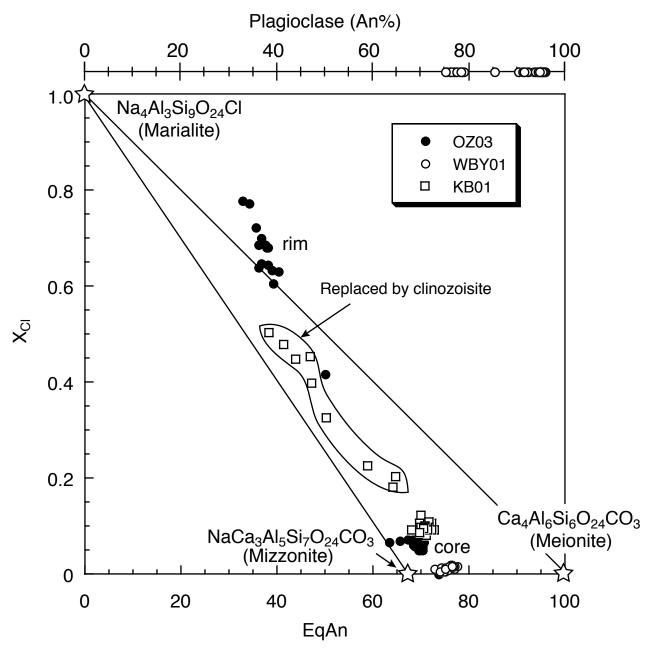


FIGURE 8. Compositional range of scapolite and plagioclase in calc-silicate-rock from the Mogok metamorphic belt in EqAn [= (AI-3)/3 X 100] –XCI [= CI/2 for total cations =  $16 \approx$  CI/(CI + CO3)] diagram. Lines bracket the stoichiometric range of scapolite grains proposed by Evans, Shaw & Haughton (1969).

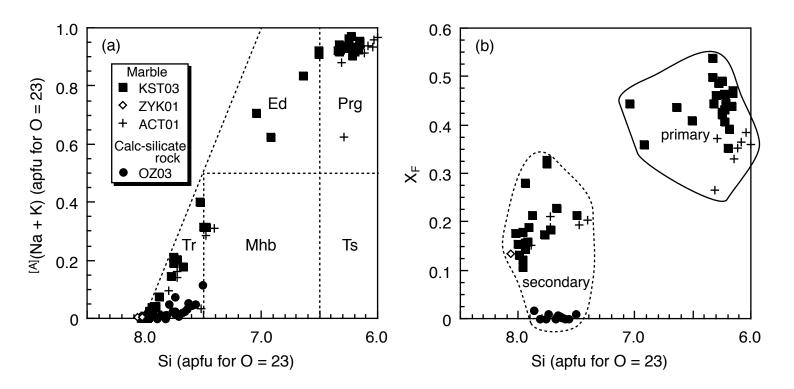


FIGURE 9. Compositional range of amphibole in marbles and calc-silicate rock from the Mogok metamorphic belt in (a)  $Si^{[A]}(Na + K)$  and (b)  $Si^{-}X_{F} = F/2 \approx F/(F + CI + OH)$ ] diagrams.

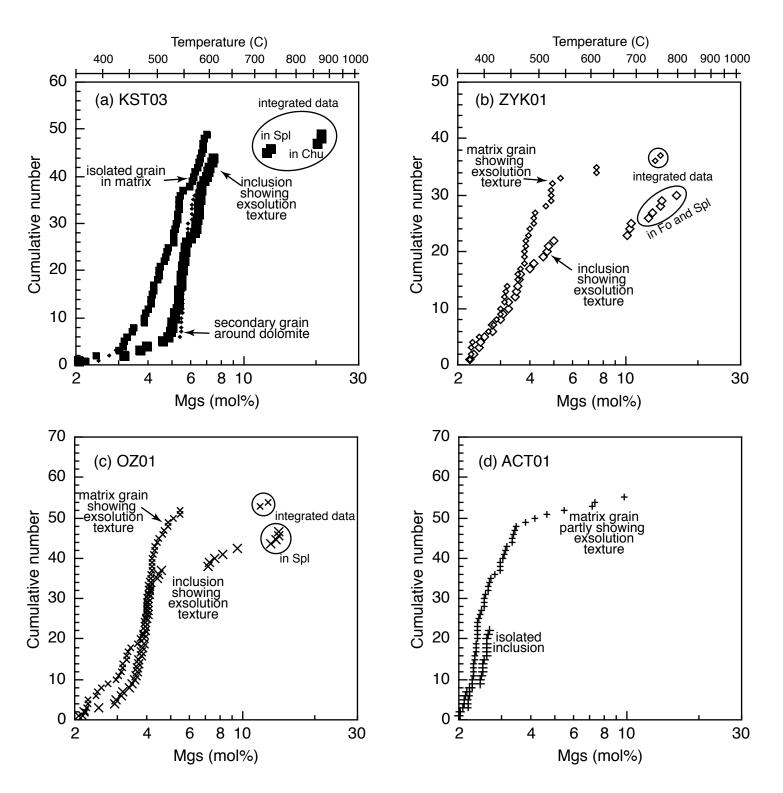


FIGURE 10. Cumulative diagrams of magnesite mol% of calcite in marble from the Mogok metamorphic belt. Data showing magnesite content less than 2 mol% are omitted. Each integrated composition was estimated using dataset of a whole inclusion or core part of an exsolved matrix grain. Temperature conditions were estimated using a geothermometer proposed by Anovitz and Essene (1987).

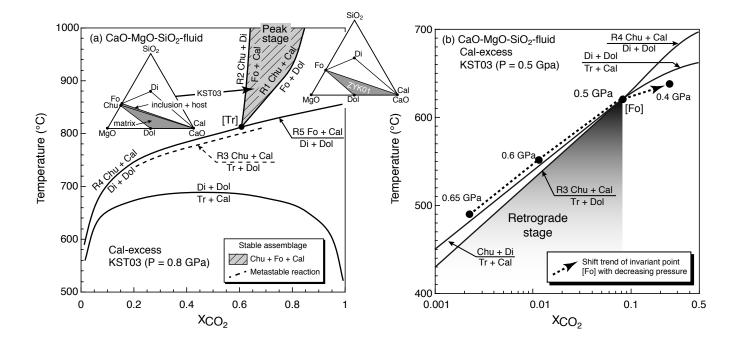


FIGURE 11. Isobaric T–XCO2 diagrams in the calcite-excess system for the clinohumite-bearing marble of KST03 from the Mogok metamorphic belt at pressures of (a) 0.8 GPa and (b) 0.5 GPa. Fixed activities of clinohumite at temperatures of 800 °C and 600 °C were employed for calculations of the diagrams (a) and (b), respectively. The pressure dependence of the upper stability limit of XCO2 and temperature conditions of the reaction R3 Tr + 13Dol + H2O = 2Chu + 15Cal + 11CO2 is shown in (b) for the pressure range of 0.4–0.65 GPa. Labels R1–R5 correspond to reactions discussed in the text. H2O and CO2 components of reactions are omitted for convenience. Minerals other than quartz, which are in equilibrium with calcite, are shown in CaO–MgO–SiO2 diagrams.

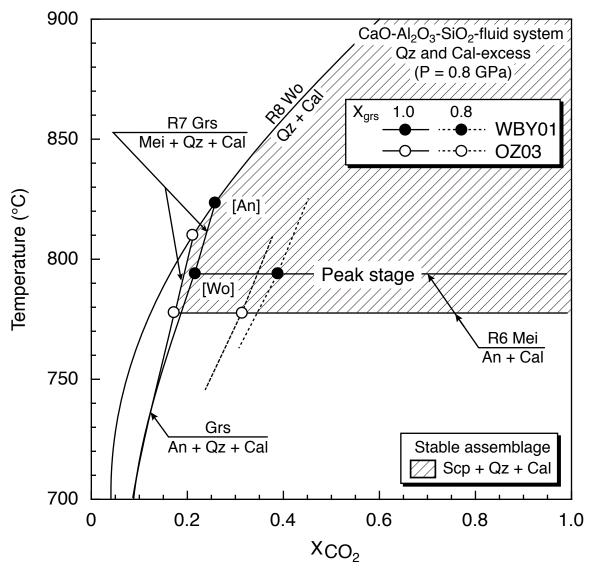


FIGURE 12. Isobaric T–XCO2 diagram in the quartz and calcite-excess system for the scapolite-bearing calc-silicate rock of WBY01 and OZ03 from the Mogok metamorphic belt showing stability relationship of scapolite and related phases at P = 0.8 GPa. Fixed activities of meionite and anorthite at temperatures of 800 °C, and that of grossular calculated as (XGrs)3, assuming ideal solid solution (where XGrs implies molecular proportion of grossular), were employed for calculation of the diagrams. H2O and CO2 components of reactions are omitted for convenience. Labels R6–R8 correspond to those of the reactions discussed in the text.

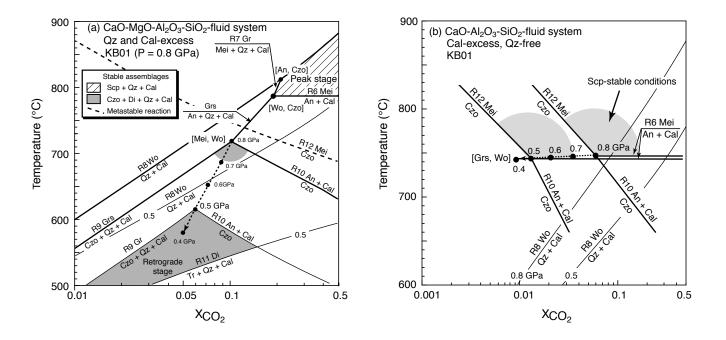


FIGURE 13. Isobaric T–XCO2 diagrams for the scapolite-bearing calc-silicate rock of KB01 from the Mogok metamorphic belt showing stability relationships of scapolite and related phases (a) in the quartz and calcite-excess system at P = 0.8 GPa and (b) in the quartz-free and calcite-excess system under various pressure conditions between 0.4 and 0.8 GPa. Univariant lines of Wo = Qz + Cal at 0.5 and 0.8 GPa are superimposed on the diagram (b) to compare the stability conditions of Qz + Cal and Czo. Fixed activities of phases at temperatures of 800 °C and 750 °C were employed for calculations of the diagrams (a) and (b), respectively. H<sub>2</sub>O and CO<sub>2</sub> components of reactions are omitted for convenience. Labels R6–R12 correspond to those of the reactions discussed in the text. Broken lines with arrow in (a) and (b) indicate shift trend of invariant points [Mei, Wo] and [Grs, Wo], respectively.

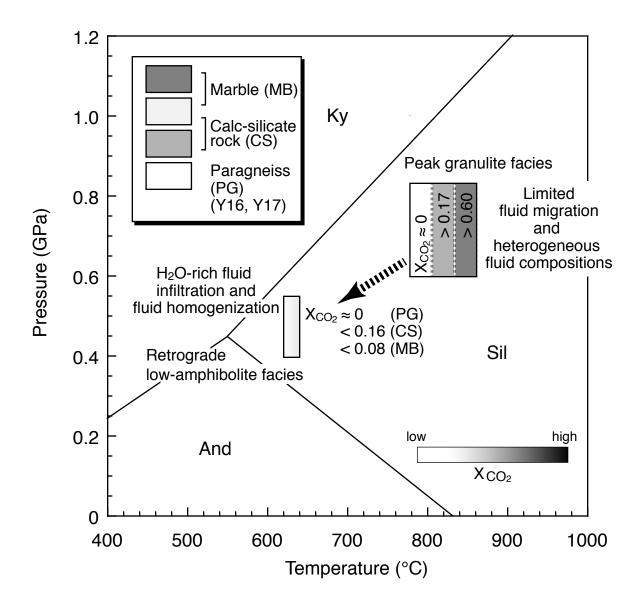


FIGURE 14. Schematic diagram showing evolution of fluid compositions of marble, calc-silicate rock, and paragneiss from the middle segment of the Mogok metamorphic belt in Myanmar. The stabilities of aluminum silicates are from Pattison (2001). References for P-T estimations of paragneiss are Y16, Ye Kyaw Thu et al. (2016); Y17, Ye Kyaw Thu et al. (2017).

Sample	Cal	Dol	Chu	Spl	Fo	Di	Phl	Prg/Ed	Tr	Scp	Pl	Qz	Czo	Ttn	Others	Note
Marble																
KST03	+, i	is, ei	+	+, i	i	r	+	+	r						Gr/Chl	1)
ZYK01	+, i	is, ei, em		+	+		+		r						Srp	
OZ01	+, i	ce, ei, em		+			+								Chl	
ACT01	+, i	em				+	+	+	r						Gr	2)
Calc-silica	ate rock															
OZ03	+, i					+			r	+, i, r		+		+, i	Kfs	
WBY01	+, r					+				+	+, r	+		+	Kfs/Ms	
KB01	+					+				+, r		+	r	+, i	Kfs	

 $r_{1}$   $r_{2}$   $r_{3}$   $r_{4}$   $r_{5}$   $r_{1}$   $r_{1$ 

Sample	labelikkyli		t the Mogo	SK IIICUIIK	KST03	t, 1 <b>v</b> 1 y amma	1.		
Mineral	Chu	Chu	Fo	Phl	Phl	Spl	Prg	Tr	Di
Note	Pgp	Pgp	Pgp	Pgp	Pgp	Pgp	Pgp	symp	symp
wt%									
SiO <sub>2</sub>	38.27	38.44	43.40	38.03	38.90	0.00	45.06	59.34	55.62
$TiO_2$	0.06	0.04	0.00	0.30	0.28	0.03	0.14	0.04	0.01
$Al_2O_3$	0.01	0.00	0.01	16.29	15.23	70.19	14.24	0.20	0.28
$Cr_2O_3$	0.00	0.00	0.00	0.00	0.00	0.01	0.07	0.02	0.00
$V_2O_3$						1.21			
FeO*	0.25	0.22	0.40	0.02	0.03	0.40	0.10	0.04	0.09
MnO	0.00	0.00	0.00	0.00	0.02	0.03	0.00	0.00	0.00
MgO	57.36	58.16	57.57	26.63	27.10	28.00	20.66	24.54	18.66
NiO			0.01						
ZnO						0.39			
BaO				7.86	7.40	0.00			
CaO	0.02	0.02	0.01	0.05	0.05	0.00	12.94	13.80	25.34
Na <sub>2</sub> O	0.00	0.00	0.00	1.02	0.98	0.00	3.35	0.09	0.03
K <sub>2</sub> O	0.00	0.00	0.00	6.18	6.43		0.22	0.05	0.01
F	4.14	3.62		3.37	3.63		2.47	0.73	
Cl	0.00	0.01		0.06	0.06		0.06	0.01	
O=F, Cl	1.74	1.53		1.43	1.54		1.05	0.31	
Total	98.37	98.98	101.40	98.38	98.57	100.26	98.26	98.55	100.04
Formula									
0	17	17	4	11	11	4	23	23	6
Si	4.01	3.99	1.00	2.73	2.79	0.00	6.33	8.00	2.00
Ti	0.01	0.00	0.00	0.02	0.02	0.00	0.02	0.00	0.00
Al	0.00	0.00	0.00	1.38	1.29	1.97	2.36	0.03	0.01
Cr	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.00
V						0.02			
Fe <sup>3+**</sup>						0.01	0.01	0.00	
$Fe^{2+**}$	0.02	0.02	0.01	0.00	0.00	0.00	0.01	0.01	0.00
Mn	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Mg	8.95	8.99	1.98	2.85	2.89	0.99	4.33	4.93	1.00
Ni			0.00						0.00
Zn						0.01			0.00
Ba	0.00	0.00		0.22	0.21	0.00			0.00
Ca	0.00	0.00	0.00	0.00	0.00	0.00	1.95	1.99	0.98
Na	0.00	0.00	0.00	0.14	0.14	0.00	0.91	0.02	0.00
Κ	0.00	0.00	0.00	0.57	0.59		0.04	0.01	0.00
Total	12.99	13.01	3.00	7.92	7.92	3.00	15.95	15.00	3.99
F	1.37	1.19		0.77	0.82		1.10	0.31	
Cl	0.00	0.00		0.01	0.01		0.01	0.00	

Table 2. Representative chemical compositions of major constituent minerals of marble from the Onzon-Thabeikkyin region of the Mogok metamorphic belt, Myanmar.

\* Total iron as FeO.

\*\* Calculated values.

Abbreviations for texture: Pgp, prograde phase; Rgp, retrograde phase; Symp, retrograde symplectitic phase.

Table 2 (continued).										
Sample	ZYK01			OZ		ACT01				
Mineral	Fo	Phl	Spl	Tr	Phl	Spl	Phl	Di	Prg	Tr
Note	Pgp	Pgp	Pgp	Rgp	Pgp	Pgp	Pgp	Pgp	Pgp	Rgp
wt%	10 50	44	0.00	50.00	10.00	0.01	44.00	<b>F1</b> 10	10 15	
SiO <sub>2</sub>	42.50	41.70	0.03	59.03	40.28	0.01	41.80	51.43	42.45	58.61
TiO <sub>2</sub>	0.00	0.16	0.06	0.04	0.86	0.04	0.20	0.35	0.22	0.05
$Al_2O_3$	0.00	16.59	71.17	0.09	16.72	70.01	14.89	5.18	17.41	1.54
$Cr_2O_3$	0.02	0.00	0.00	0.01	0.00	0.00	0.00	0.01	0.06	0.01
$V_2O_3$			0.10			0.05				
FeO*	0.69	0.14	0.77	0.14	0.41	1.91	1.29	2.02	2.05	1.73
MnO	0.02	0.01	0.02	0.01	0.00	0.01	0.01	0.04	0.01	0.00
MgO	56.33	26.75	26.99	24.90	25.84	25.53	26.51	15.42	18.23	22.94
NiO	0.04									
ZnO	0.00	0.00	0.16		0.02	1.96	0.17	0.00		
BaO	_	0.09	_		0.16	_	0.19			
CaO	0.00	0.01	0.00	13.92	0.04	0.00	0.05	24.82	13.06	13.38
Na <sub>2</sub> O	0.00	0.91	0.00	0.08	0.33	0.00	0.32	0.34	2.30	0.29
K <sub>2</sub> O	0.00	8.88	0.00	0.03	10.26	0.00	10.13	0.00	1.99	0.08
F		1.68		0.62	2.12		2.45		1.63	0.71
Cl		0.03		0.02	0.02		0.06		0.06	0.00
O=F, Cl		0.71		0.27	0.90		1.05		0.70	0.30
Total	99.60	96.24	99.30	98.62	96.16	99.52	97.02	99.61	98.77	99.04
Formula										
0	4	11	4	23	11	4	11	6	23	23
Si	1.00	2.88	0.00	7.96	2.82	0.00	2.91	1.88	6.00	7.91
Ti	0.00	0.01	0.00	0.00	0.05	0.00	0.01	0.01	0.02	0.01
Al	0.00	1.35	2.01	0.01	1.38	2.00	1.22	0.22	2.90	0.25
Cr	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.00
V			0.00			0.00				
Fe <sup>3+**</sup>			0.00	0.01		0.00			0.09	0.00
Fe <sup>2+**</sup>	0.01	0.01	0.02	0.00	0.02	0.04	0.08	0.06	0.16	0.20
Mn	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Mg	1.98	2.75	0.96	5.00	2.69	0.92	2.75	0.84	3.84	4.62
Ni	0.00									
Zn	0.00	0.00	0.00		0.00	0.04	0.01	0.00		
Ba		0.00			0.00		0.01			
Ca	0.00	0.00	0.00	2.01	0.00	0.00	0.00	0.97	1.98	1.94
Na	0.00	0.12	0.00	0.02	0.05	0.00	0.04	0.02	0.63	0.08
Κ	0.00	0.78	0.00	0.01	0.92	0.00	0.90	0.00	0.36	0.01
Total	3.00	7.90	2.99	15.04	7.93	3.00	7.94	4.01	15.98	15.00
F		0.37		0.26	0.47		0.54		0.73	0.30
Cl		0.00		0.00	0.00		0.01		0.01	0.00

Table 2 (continued).

Sample							WBY01	KB01				
Mineral	Scp	Scp	Di	Tr	Scp	P1	P1	Di	Ms	Scp	Scp	Czo
	core	mantle	Pgp	Rgp	Pgp	Pgp	Rgp	Pgp	Rgp	Pgp	Rgp	Rgp
Note	(Pgp)	(Rgp)	1 SP	Rsp	1 SP	1 SP	Rsp	1 SP	кsр	1 SP	Кър	кsр
wt%												
SiO <sub>2</sub>	45.21	54.62	52.62	54.64	43.13	44.27	49.55	52.66	46.75	44.17	51.62	38.74
TiO <sub>2</sub>	0.01	0.00	0.16	0.17	0.03	0.00	0.00	0.07	0.02	0.00	0.02	0.05
$Al_2O_3$	27.98	23.83	1.81	2.10	29.15	35.46	32.50	0.94	33.10	28.36	25.5	27.76
$Cr_2O_3$	0.00	0.00	0.06	0.06	0.03	0.00	0.03	0.03	0.00	0.00	0.00	0.02
FeO*	0.15	0.00	8.10	10.08	0.12	0.04	0.02	10.35	1.64	0.09	0.07	7.41**
MnO	0.00	0.00	0.13	0.06	0.01	0.00	0.00	0.31	0.00	0.00	0.01	0.02
MgO	0.06	0.00	12.93	17.56	0.04	0.00	0.00	11.33	2.55	0.07	0.01	0.05
ZnO									0.09	0.00	0.00	0.00
BaO									0.00			
CaO	19.07	9.62	23.91	12.64	20.72	19.47	15.62	24.70	0.07	19.52	12.37	23.80
Na <sub>2</sub> O	2.80	8.09	0.31	0.20	1.75	0.45	2.74	0.12	0.18	2.34	6.49	0.00
K <sub>2</sub> O	0.24	0.73	0.00	0.07	0.15	0.02	0.08	0.02	10.90	0.41	0.58	0.00
F	0.06	0.00		0.00	0.03				0.07	0.00	0.00	0.00
Cl	0.22	2.63		0.00	0.07				0.00	0.36	1.61	0.00
O=F, Cl	0.07	0.59		0.00	0.03				0.03	0.08	0.36	0.00
Total	95.73	98.93	100.03	97.58	95.20	99.76	100.56	100.76	95.40	95.24	97.92	90.44
Formula												
0	16†	16†	6	23	16†	8	8	6	11	16†	16†	12.5
Si	6.92	7.97	1.96	7.75	6.67	2.05	2.25	1.98	3.13	6.81	7.62	3.02
Ti	0.00	0.00	0.00	0.02	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Al	5.05	4.10	0.08	0.35	5.31	1.94	1.74	0.04	2.61	5.16	4.44	2.55
Cr	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
$Fe^{3+}$				0.13 <sup>§</sup>								0.43**
Fe <sup>2+</sup> *	0.02	0.00	0.25	1.07 <sup>§</sup>	0.02	0.00	0.00	0.33	0.09	0.01	0.01	0.00
Mn	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.00
Mg	0.01	0.00	0.72	3.71	0.01	0.00	0.00	0.64	0.25	0.02	0.00	0.01
Zn									0.00	0.00	0.00	0.00
Ba	2 1 2	1.50	0.00	1.02	2 42	0.07	0.76	0.00	0.00	2.02	1.00	1.00
Ca Na	3.13 0.83	1.50 2.29	0.96 0.02	1.92 0.06	3.43 0.52	0.97 0.04	0.76 0.24	0.99 0.01	0.01 0.02	3.23 0.70	1.96 1.86	1.99 0.00
Na K	0.85	0.14	0.02	0.08	0.52	0.04	0.24	0.01	0.02	0.70	0.11	0.00
r Total	16.00	16.00	4.00	13.83	16.00	5.00	5.00	4.00	0.93 7.04	0.08 16.00	16.00	0.00 7.56
TOTAL	10.00	10.00	00	15.05	10.00	5.00	5.00	т.00	7.04	10.00	10.00	1.50
F	0.03	0.00		0.00	0.02				0.02	0.00	0.00	0.00
Cl	0.06	0.65		0.00	0.02				0.00	0.09	0.40	0.00

Table 3. Representative chemical compositions of major constituent minerals of calc-silicate rock from the Onzon-Thabeikkyin region of the Mogok metamorphic belt, Myanmar.

\* Total iron as FeO.

\*\* Total iron as Fe<sub>2</sub>O<sub>3</sub>.

† Total cations as 16.

§ Calculated value (see text).

Abbreviations for texture: Pgp, prograde phase; Rgp, retrograde phase.

Sample	Chu	Tr		Di	Scp		P1		Czo	Figure
	$X_F$	$X_{OH}$	$X_{Mg}$	$X_{Mg}$	EqAn		An		$X_{Fe}$	
	Pgp	Pgp	Rgp	Pgp	Pgp	Rgp	Pgp	Rgp	Rgp	
KST03	0.63	0.80		1.00						Fig. 11
OZ03					68		82*			Fig, 12
WBY01					78		95			Fig, 12
KB01			0.80**	0.80	70		85*		0.15	Fig. 13a
KB01						65		78*	0.15	Fig. 13b

Table 4. Chemical compositions of minerals employed for the calculations of  $T-X_{C02}$  diagrams.

\* Anorthite content was estimated assuming that An/EqAn = 1.2 considering the pair of prograde scapolite and plagioclase in WBY01.

\*\*  $X_{Mg}$  value was assumed to be same with that of diopside.

Abbreviations for texture: Pgp, prograde phase; Rgp, retrograde phase.