

Petrological implication of wagnerite-*Ma5bc* in the quartzofeldspathic gneiss, Larsemann Hills, East Antarctica^{*}

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Abstract Wagnerite-*Ma5bc* polytype is found in a gneiss associated with cordierite-prismatine-bearing gneiss, northern Stomes Peninsula, Larsemann Hills, East Antarctica. Wagnerite coexists with apatite and both of them are present as rock-forming minerals. Ca depletion is not essential to the formation of wagnerite; sufficient Mg, P and F components, and a relatively low Ca/Mg ratio is critical; otherwise only apatite crystallizes. Strong anatexis of the quartzofeldspathic gneiss of the area results in differentiation and separate enrichment of leuco- and melanocratic components. P and F tend to accumulate in the mafic-rich melanocratic component, and when their abundance reaches certain concentration, wagnerite forms. It is perhaps the chemistry of the setting that controls the appearance of wagnerite, whereas the *PT* condition is responsible for the variety of wagnerite polytype. A disordered polytype crystallizes at relatively high temperature, then cools down slowly and transforms into one of the ordered polytypes, in this case, the *Ma5bc* polytype.

Keywords: wagnerite-*Ma5bc* polytype, anatexis, formation condition, volatile components, Larsemann Hills

Mg-dominant minerals of wagnerite-triplite group, i.e. $(\text{Mg, Fe, Mn})_2(\text{PO}_4)(\text{F, OH})$, are relatively rare, but have been found in a wide variety of geologic environments, including metamorphic rocks of all grades^[1] such as quartz-carbonate veins of hydrothermal origin, pegmatites, kyanite-quartzite, mica schist, sillimanite gneiss, and magnetite ores.

Up to now, wagnerite is present mainly as the *Ma2bc* polytype, an accessory mineral in calcium-poor rocks^[2,5] in which apatite, if present, often occurs as retrograde rim around wagnerite. Henriques^[6] reported a small amount of wagnerite and apatite in kyanite-quartzite, but the relation between the two minerals was not mentioned. The wagnerite-bearing rocks generally have other phosphate minerals like monazite and xenotime.

The polytype with a 5-fold *b*-dimension repeat was first discovered on Stomes Peninsula in the Larsemann Hills, East Antarctica^[7,8]. The Larsemann Hills rock is distinctive in that wagnerite coexists with apatite and both are present as major rock-forming constituents, which has not been reported before. This paper focuses on the petrological implications and geological setting of the wagnerite-apatite

assemblage in the study area.

1 Regional geology

The Larsemann Hills were once situated at the East Antarctic craton of the Gondwana supercontinent, where the *c.* 550 Ma Pan-African tectonothermal event was intensively developed^[9]. A suite of high amphibolite-granulite facies rocks is present in this region, and among them the sillimanite-garnet gneisses, and the quartzofeldspathic gneisses are the main types, which are intercalated with pyroxene-biotite-plagioclase gneiss, biotite-plagioclase gneiss, minor granitic gneiss, hornblende-two pyroxene granulite, and calc-silicates. In addition, some granites and pegmatites are also present.

The gneiss containing wagnerite-*Ma5bc* occurs in the core of what appears to be a fold in banded cordierite-prismatine gneiss (Fig. 1). Prismatine and cordierite also form coarse-grained segregations and nodules associated with quartzofeldspathic segregations. Given the presence of Al-rich minerals cordierite and prismatine, the gneisses illustrated in Fig. 1 most likely have sedimentary precursors that were subsequently modified by migmatization.

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The gneiss lacks obvious foliation. Major constituents are plagioclase, wagnerite-*Ma5bc*, apatite and magnetite; subordinate are biotite, quartz, potassium feldspar, cordierite, minerals of the monazite and xenotime groups, corundum, lamellar hematite-ilmenite intergrowths, hercynite, and sulfide.

The conditions for the metamorphic peak granulite facies in the Larsemann Hills are 750 to -860°C at 0.6–0.7 GPa^[10–13].

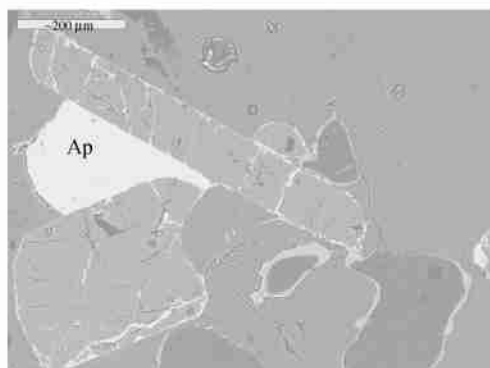


Fig. 1. BSE image of Wag-Ap and late Kfs and Ap₂.

2 Petrography and texture feature

Generally situated between mineral grains such as plagioclase, wagnerite has a straight contact with apatite (Fig. 1), which suggests the essentially co-existing feature of the two minerals. Wagnerite locally occupying the position of apatite implies the earlier beginning of apatite and late finishing of wagnerite. Following apatite and wagnerite are monazite and xenotime in small amount, and between them are basically co-existing relationships. Meanwhile, opaque minerals like magnetite, ilmenite and hematite are formed, and their volume in the rock can reach 15%.

Wagnerite-*Ma5bc* forms anhedral to euhedral grains mostly 0.5–2 mm across (the maximum is about 2.5 mm). Some grains have a tabular habit and the thickness can be less than 0.1 mm; a few of them show two terminal faces. Wagnerite-*Ma5bc* is commonly dusty with fine inclusions or discolored by incipient alteration. Textures suggest two generations of apatite: relatively coarse grains with aligned acicular inclusions; rare margins fringing wagnerite-*Ma5bc* (Fig. 1) or overgrowths separating wagnerite-*Ma5bc* from oxide and sulfide inclusions. Microscopic seams of apatite have also developed along

cracks and grain boundaries of wagnerite-*Ma5bc* (Fig. 1). Symplectitic intergrowths of cordierite with K-feldspar were developed (Fig. 2), which may correspond to the sapphirine-orthopyroxene symplectite after wagnerite in Eastern Ghats Land, India^[14]. In summary, textures are consistent with a primary assemblage: wagnerite-*Ma5bc* + plagioclase + apatite(1) + magnetite + ilmenite-hematite.

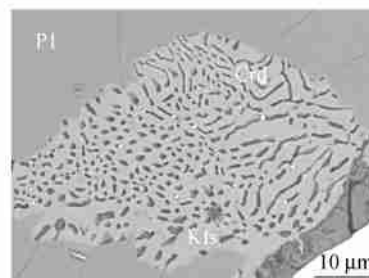
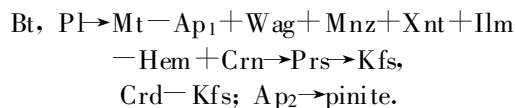


Fig. 2. BSE image of Kfs-Crd symplectite after wagnerite (the background is Kfs).

Although interlayered with cordierite-prismatine gneiss the wag-bearing gneiss has not the contact texture between wag and prs. By combining the relatively earlier crystallization of wag than monazite-xenotime in the gneiss while in the prs gneiss mnz-xnt formed earlier, the possible mineral crystallization sequence relative to wag can be summarized as follows (mineral abbreviations are after Kretz^[28]):



Predating prismatine and the crd-kfs symplectite, the formation of wagnerite is related to the biotite dehydration reaction with plagioclase involved in anatexis, and should be the stage of garnet in quartzofeldspathic gneisses, that is, wagnerite formed at the peak metamorphism.

Semi-quantitative energy spectrum composition scanning demonstrates that from Ap₁ to Ap₂, Mg and P tend to increase, while Na, Ca, Fe, and S tend to decrease.

3 Crystal structure of wagnerite

By using a Bruker Smart Apex CCD system and graphite-monochromated MoK α radiation, the crystal structure of wagnerite is measured and the unit-cell parameters were obtained by least-squares refinement, $a = 9.645(2) \text{ \AA}$, $b = 31.659(6) \text{ \AA}$, $c = 11.914(2) \text{ \AA}$, $\beta = 108.26(3)^{\circ}$, $V = 3455(1) \text{ \AA}^3$,

and $Z=40$. The wagnerite- $Ma5bc$ ^[7,8], space group Ia , distinct to that of the type wagnerite, $2b$ polytype (space group $P2/a$), is a new polytype. The primary difference between the wagnerite- $Ma5bc$ and wagnerite- $Ma2bc$ is ordering of the (F, OH) positions. F can occupy one of the two positions, which can result in two distinct configurations along the a direction. Other polytypes have been summarized by Chopin et al.^[15].

4 Mineral chemistry

Microprobe analyses of wagnerite are given in Table 1. Assuming Fe, Ti, Ca, and Mn substitute for Mg, all the spots (except spots 1-1 and 3-1) give the average composition as follows: $(Mg_{19.050}Fe_{1.006}Ti_{0.192}Ca_{0.030}Mn_{0.023})_{20.301}(P_{9.982}Si_{0.018})_{10.000}O_{40}F_{6.485}(OH)_{3.545}$; the presence of hydroxyl was confirmed by Raman spectroscopy.

Table 1. Microprobe analyses of wagnerite^{a)}

Oxides	1-1	1-2	2-2	2-3	2-5	2-6	2-7	3-1	3-2	3-3	4-1	8-4	8-2
MgO	43.59	44.13	44.99	45.15	44.29	45.05	45.07	42.59	44.45	43.85	44.80	44.85	45.03
SiO ₂	0.064	0.056	0.061	0.077	0.053	0.027	0.080	0.051	0.057	0.053	0.084	0.072	0.075
CaO	0.088	0.105	0.081	0.084	0.071	0.075	0.080	2.374	0.089	0.097	0.084	0.129	0.158
TiO ₂	0.974	0.954	0.851	0.826	0.821	0.745	0.722	0.975	1.028	0.949	0.974	1.007	0.924
MnO	0.084	0.096	0.088	0.058	0.095	0.078	0.106	0.089	0.132	0.121	0.065	0.088	0.12
FeO	4.153	4.809	4.236	4.100	4.610	2.563	4.051	4.314	4.299	4.774	3.959	4.457	4.431
P ₂ O ₅	41.73	41.72	41.41	40.95	41.47	41.37	41.08	40.36	41.27	41.06	41.80	41.2	40.38
F	6.198	6.643	6.470	7.681	7.732	6.978	6.734	6.731	7.101	6.298	6.833	8.801	7.605
Cl												0.003	0.001
Total	94.30	95.72	95.49	95.70	95.91	93.98	95.09	94.70	95.44	94.56	95.73	96.93	95.52
OH	5.546	5.944	5.790	6.872	6.918	6.243	6.025	6.022	6.354	5.635	6.114	2.428	3.574
Mg	18.390	18.626	19.128	19.414	18.804	19.178	19.318	18.580	18.965	18.807	18.887	19.168	19.634
Si	0.018	0.015	0.017	0.022	0.015	0.008	0.023	0.014	0.016	0.015	0.024	0.020	0.022
Ca	0.027	0.032	0.025	0.026	0.022	0.023	0.025	0.744	0.027	0.030	0.026	0.040	0.050
Ti	0.207	0.203	0.183	0.179	0.176	0.160	0.156	0.215	0.221	0.205	0.207	0.217	0.204
Mn	0.020	0.023	0.021	0.014	0.023	0.019	0.026	0.022	0.032	0.030	0.016	0.021	0.030
Fe	0.983	1.139	1.010	0.989	1.098	0.612	0.974	1.056	1.029	1.149	0.936	1.069	1.084
F	5.548	5.949	5.836	7.007	6.965	6.302	6.123	6.230	6.428	5.730	6.106	7.980	7.035
OH	4.452	4.051	4.164	2.993	3.036	3.699	3.877	3.770	3.573	4.270	3.894	2.201	3.306
Cl												0.001	0.001

a) Average of 9 electron microprobe analyses JXA-8800A, 20 kV, 20 nA, 5–20 μm, wavelength dispersion. Standard samples: apatite (P, F), wollastonite (Si, Ca), rutile (Ti), hematite (Fe), Mn oxide (Mn), forsterite (Mg). All Fe are as FeO. Na, Al, K, and Cr are below detection. H₂O are calculated assuming F+OH=10; Cations and F anions calculations are based on the assumption of 10 (PO₄)³⁻. The too low MgO contents of spot 1-1 may result from systematic error, while the too high CaO and low MgO of spot 3-1 may be affected by apatite near the measured spot. The measurement was carried out at the Electron Microprobe Analysis Laboratory of the Institute of Mineral Resources, Chinese Academy of Geological Sciences.

There is a positive correlation between Ti and Fe, $Ti \propto Fe$, but $(Fe+Ti)$ and Mg show negative correlation (Fig. 3). $(Fe+Ti)$ as a whole replaces Mg. There is a positive correlation between F and Mg (Fig. 4), which is consistent with the Mg to F and Fe to Cl rule^[17]. Of course, both F and Mg show negative correlation with $(Fe+Ti)$.

The associated minerals, notably feldspar, have measurable phosphorus contents (Table 2). Through measurements of many spots (for a short text only two spots are listed in Table 2), the changes between grains result from the possible substitute styles:

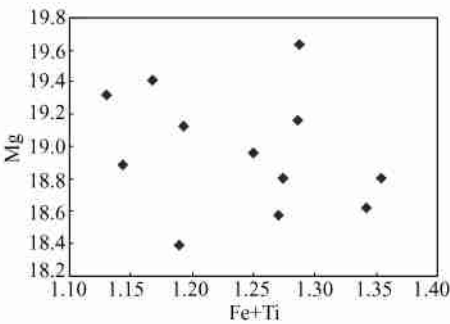


Fig. 3. $Fe+Ti$ versus Mg contents in wagnerite formula (per 10P).

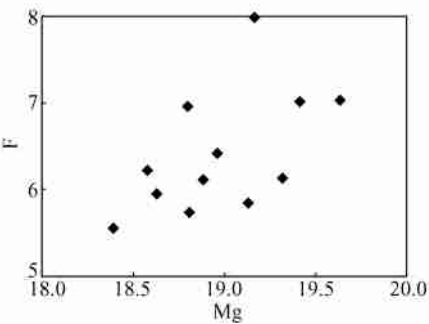
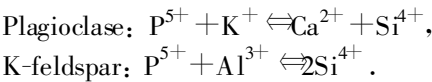


Fig. 4. F versus Mg contents in wagnerite fomula (per 10P).



The substitution in plagioclase abides by the charge balance rule, while in K-feldspar, structure (ion groups) can maintain balance in addition, thus the substitute is easier and the amount of phosphorus in Kfs is larger (Table 2).

Late biotite has lower F and higher Cl than early biotite, but Ti contents are about the same for both (Table 2). Chlorine/fluorine ratio is higher in apatite than coexisting biotite.

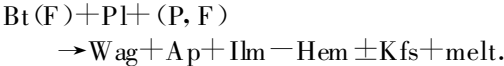
Table 2. Microprobe analyses of minerals associated with wagnerite^{a)}

Oxides	8-2-ap1	8-3-ap1	8-1-pl	8-5-pl	8-3-1kfs	8-3-2kfs	8-4-bt1	8-2-bt1	8-5-bt2
MgO	0.311	0.28	0.019	0	0.068	0	15.879	15.467	15.076
SiO ₂	0	0	63.024	62.497	60.78	64.824	36.437	35.722	36.437
Al ₂ O ₃	0	0	21.379	21.882	18.856	18.793	14.022	13.818	13.912
CaO	50.566	48.91	3.406	3.693	0.068	0.009	0	0.017	0.017
TiO ₂	0	0	0.014	0	0.024	0.017	4.848	5.365	5.991
MnO	0.198	0.183	0.004	0.006	0	0	0	0.034	0
FeO	1.489	1.913	0.059	0.059	0.412	0.128	12.948	13.076	13.328
Cr ₂ O ₃	0.008	0.015	0.018	0.016	0	0	0.017	0.014	0.027
Na ₂ O	0.268	0.511	9.056	8.934	0.524	0.252	0.032	0.097	0.068
K ₂ O	0	0.01	1.02	0.703	14.592	16.766	10.64	9.516	10.362
P ₂ O ₅	41.044	40.257	0.404	0.325	0.362	0.912	0.053	0.065	0.018
F	2.335	2.811	0	0.09	0	0.04	1.234	0.987	0.622
Cl	1.925	2.087	0.002	0	0.069	0.003	0.481	0.364	0.505
Total	96.727	95.322	98.405	98.167	95.739	101.726	95.962	94.044	95.987
Mg	0.040	0.037	0.001	0	0.005	0	1.78	1.759	1.684
Si	0	0	2.845	2.826	2.939	2.975	2.739	2.725	2.73
Al	0	0	1.138	1.166	1.075	1.016	1.242	1.242	1.228
Ca	4.746	4.674	0.165	0.179	0.004	0	0	0.002	0.002
Ti	0	0	0	0	0.001	0.001	0.274	0.308	0.337
Mn	0.015	0.013	0	0	0	0	0	0.002	0
Fe	0.109	0.143	0.002	0.002	0.017	0.005	0.814	0.834	0.835
Cr	0.001	0.002	0.001	0.001	0	0	0.001	0.001	0.002
Na	0.046	0.088	0.793	0.783	0.049	0.022	0.004	0.015	0.01
K	0	0.001	0.059	0.04	0.9	0.982	1.02	0.926	0.99
P	3.043	3.040	0.015	0.013	0.015	0.035	0.003	0.005	0.001
Oxygen	12	12	8	8	8	8	23	23	23
F	0.648	0.793	0	0.013	0	0.006	0.292	0.239	0.147
Cl	0.287	0.316	0	0	0.006	0	0.061	0.047	0.065
An			0.172	0.186					
Cl/(F+Cl)	0.306	0.285					0.173	0.165	0.303
Mg/(Mg+Fe)							0.686	0.678	0.668

a) Analysis condition is the same as that of Table 1.

5 Formation process

According to the petrographic observation and analysis, wagnerite and apatite formation might be due to the introduction of P and F during the incongruent partial melting of biotite and plagioclase at the peak of granulite facies metamorphism.



Here HIM represents the synneusis assemblage of hematite-ilmenite-magnetite.

However, the abundance of phosphate minerals in metamorphic rocks throughout Stomes Peninsula

suggests the possibility that the sedimentary precursors are originally enriched in P, and introduction of P need not be invoked to explain the formation of apatite and wagnerite. In the following subsections we will discuss the factors relating to the formation of wagnerite.

5.1 Volatiles

According to the affinity feature to fluids and melts, the solubility of the volatile component F in melt is proportional to the atom radius of alkaline earth elements, i. e. F prefers to form valence with larger alkaline earth elements in melt, while Cl is favorable to concentrate in fluid phase^[18].

The volatiles in wagnerite in the study area is basically F, with small amount of OH (Table 1), which suggests the low pressure setting^[19]. Volatile components in apatite, too, are predominantly F and some Cl (Table 2). Earlier biotite involved in anatexis has more F, while the late biotite after anatexis has more Cl. The petrographic feature suggests wagnerite formed at the intensive period of anatexis. Due to the decomposition of biotite and migration of components, F, P accompanying Mg(Fe) concentrate at a certain position which may be favorable to the formation of wagnerite. The subhedral to euhedral crystal shapes of the mineral show that there is a free space, such as melt or solution media setting, for wagnerite crystallization.

In the general geological setting, the fluids with temperature under typical magmatic condition may have the forms like melt and solution, such as aluminum silicate melt, aqueous salt melt and hydrothermal dilute fluid (H₂O, CO₂ predominant)^[20]. The aqueous salt melt includes the phosphate and borate of alkaline and alkaline earth metals and their fluorine and chlorine compounds, and considerable amount of SiO₂ and Al₂O₃. The formation condition of wagnerite of the study area may be close to the setting of aluminum silicate melt and/or aqueous salt melt.

By experiments, the F-P rich fluids are strong acid in feature and can prohibit the crystallization of feldspar, especially K-feldspar. Furthermore, the strong acid fluid can enhance the activity of SiO₂^[21, 22], which is consistent with the erosion of plagioclase and quartz around wagnerite and the late presence of K-feldspar.

5.2 Composition condition

It is generally considered that wagnerite forms under calcium-poor condition^[2-5]. According to the analysis of Simmat & Rickers^[14], if Ca/(Ca+Mg) ~ 0.02, only wagnerite occurs; if the ratio reaches ~ 0.25, the retrograde apatite rim around wagnerite can be formed. Irouschek-Zumthor and Armbruster^[2] emphasized that calcium-poor was not the prerequisite condition for the occurrence of wagnerite, the point lies in that there is too low amount of CaO components (Ca/P < 5/3) available for the formation of apatite.

For the case of the Larsemann Hills, the association of wagnerite and apatite and their relative contents suggest that the whole-rock Ca/(Ca+Mg) ratio is 0.50 or greater, i. e. the rock is not depleted in Ca, while the mobile cation components being rich in MgO and CaO. Therefore, the factors for occurrence of the phosphate association of wagnerite and apatite, and monazite and xenotime, can result in the enrichment of P and F, but short of Ca, i. e. low Ca/Mg in the media. The appearance of wagnerite could be due to the high bulk P content, contemporarily the efficient CaO component may satisfy the condition that Ca/P ratio is less than 5/3 and not all phosphate can be accommodated in apatite, otherwise only apatite would be present.

Wagnerite (*Ma5bc*) crystallized at the syn- to late stage of early apatite (Ap₁): initially the CaO-rich apatite formed, part of the CaO composition sourced from the decomposition of plagioclase, when MgO began to concentrate and wagnerite, and co-existence of Ap-Wag occurred; then CaO depleted but MgO was still abundant enough, wagnerite crystallized continuously while apatite was eroded, which suggests that the supply of the components P, F is not persistent, and Ap-Wag is formed only locally at a short period. The continual supply of MgO was demonstrated by the cordierite symplectite after wagnerite. Trace amount of late apatite (Ap₂) formed even later.

Accompanying the occurrence of wagnerite, anatexis developed intensively and granitic gneisses and granite pegmatites prevail in the Larsemann Hills. Meanwhile, the presence of a considerable amount of residual opaque or melanocratic mineral associations, such as iron-titanium oxides and prismatine-cordierite, suggests the substantial differentiation of

mafic and sialic components in the anatexis process^[23], in the former wagnerite crystallized. Phosphorus tends to concentrate in the rocks rich in MgO, FeO and TiO₂ but poor in K₂O and SiO₂^[24], because Mysen et al.^[25] pointed out that when non-bridge oxygen (NBO) is present in the melt, joining of P will decrease the amount of NBO and P tends to form complexes with cations Ca²⁺, Mg²⁺ and Na⁺.

With the joining of P₂O₅, the immiscibility scope of melts of different compositions resulting from anatexis will enlarge^[26,27], which affects the distribution of trace elements between melts; presence of P₂O₅ makes it easier for rare earth elements (REE) to enter the basic melt and form the REE³⁺ phosphate complex which is more stable in basic melt than in acid one. The co-existence of monazite and xenotime with wagnerite may be related to this point. By the same reason, alkaline metal cations prefer to enter the acid melt with P₂O₅ available.

5.3 Temperature constraints

The formation of wagnerite can be low to diagenesis stage^[28], high to eclogite facies^[3] or granulite facies metamorphism condition^[14]. A review of the occurrences of 40 wagnerite-triplite-group minerals of known polytype indicates that mineral composition plays a more important role than temperature and pressure conditions of formation in determining polytype (Chopin et al., unpublished data).

However, there appears to be a possible relationship between temperature and polytype for most magnesian compositions, i. e. $X_{\text{Mg}} = \text{Mg}/(\text{sum cations}) > 0.9$, which includes the Larsemann Hills sample^[8]. With one possible exception, magnesian wagnerite formed at low or moderate temperatures, i. e. less than $\sim 700^\circ\text{C}$, shows the *2b* polytype. Wagnerites of comparable F content and X_{Mg} formed at higher temperature shows any one of the four known ordered polytypes.

Wagnerite-*Ma2bc* is found in rocks formed over a wide range of temperature and pressures, magnesian wagnerite (*Ma2bc*) is generally formed under greenschist to amphibolite facies metamorphism conditions^[8], while the *Ma5bc* polytype and even highly ordered *Ma7bc* or *Ma9bc* occur at granulite facies metamorphism^[16,29]. The possible situation may be that in complex natural systems wagnerite could have crystallized as the disordered *Mabc* polytype at geo-

logically reasonable temperatures; the higher the initial temperature at which wagnerite crystallizes, the larger the cooling amplitude in temperature, and the more highly ordered polytype forms. For the case of the Larsemann Hills, the *Ma5bc* polytype may result from the initial *Mabc* or *Ma2bc* polytype, after a long term cooling, the initial polytype was inverted to *Ma5bc* polytype.

For a more general situation, one might consider three possible scenarios for the relationship between wagnerite polytype composition and temperature of formation:

Scenario 1. Each polytype has a stability field at some *P-T* condition, F content and X_{Mg} ratio, and the polytype at original crystallization is the one presently observed.

Scenario 2. There are only two stable polytypes: the disordered *1b* and ordered *2b* polytype; the other three polytypes are metastable. At $T < \sim 700^\circ\text{C}$, all OH-rich and magnesian compositions ($X_{\text{Mg}} > 0.9$) originally crystallize as *2b*, whereas fewer magnesian compositions crystallize as *1b*. At $T > 700^\circ\text{C}$, all F-rich compositions crystallize as the *1b* polytype. Upon cooling under natural conditions where annealing is likely, the *1b* structure is preserved in F-rich material only in the compositions richest in Fe and Mn. Intermediate compositions inverted to *5b*, whereas highly magnesian compositions inverted to any one of the 4 ordered polytypes.

Scenario 3. *5b* is a stable, high-*T* polytype in highly magnesian compositions, i. e. whether it remains *5b* or is inverted to *7b*, *9b*, or *2b* during cooling depends on the cooling history.

In each case, the temperature and pressure for formation of wagnerite, especially the former, can constrain the polytypes of wagnerite.

6 Conclusion

(1) Wagnerite-*Ma5bc* polytype is found in a gneiss associated with cordierite-prismatine-bearing gneiss northern Stornes Peninsula, Larsemann Hills East Antarctica. Wagnerite coexists with apatite and both of the minerals are present as rock-forming minerals. They were formed at the peak metamorphism period.

(2) Ca depletion is not essential to the formation

of wagnerite; sufficient Mg, P and F components, and a relatively low Ca/Mg ratio are critical, otherwise only apatite crystallizes.

(3) Strong anatexis of the quartzofeldspathic gneiss of the area results in differentiation and separate enrichment of leuco- and melanocratic components. P and F tend to be concentrated with Mg and Fe in more melanocratic layers, which are thus more favorable hosts for wagnerite.

(4) It is perhaps the chemistry of the setting that controls the appearance of wagnerite, whereas the *PT* conditions and post-metamorphic cooling history are responsible for the 5b polytype found in the study area.

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