Compositional variation of Fe-Ti oxides from the Sokli complex, northeastern Finland

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ABSTRACT: The phoscorite-carbonatite complex at Sokli, northeastern Finland, is composed of five stages of intrusions of phoscorites and carbonatites (P1-C1, P2-C2, P3-C3 phoscorites and calcite carbonatites, D4 and D5 dolomite carbonatites) which are cut by numerous lamprophyric dikes. Magnetite is ubiquitous in all constituent rock units of the complex and frequently associates with ilmenite. Most ilmenite intergrowths from the Sokli phoscorite-carbonatite complex occur as internal and/or external granules (composite type exsolutions) or sandwich type exsolution lamellae in the host magnetites. Discrete ilmenite crystals are found only in P3 and C3. On the other hand, ilmenites in the ultramafic lamprophyres occur as discrete crystals as well as trellis and sandwich type exsolution lamellae or composite type exsolutions in magnetite. Electron microprobe analyses reveal that magnetite of the Sokli complex belongs essentially to the magnetite-ulvöspinel solid solution series with a small proportion of magnesioferrite. The compositions of magnetite from the Sokli phoscorite-carbonatite complex vary systematically from stage to stage with increasing $Fe^{2+}/(Fe^{2+}+Mg)$ ratio, and decreasing Al, Mn and Ti contents. Magnetite from the ultramafic lamprophyre is characterized by large compositional ranges owing to the large amount of mantle-derived xenocrysts. Some grains are particularly high in Cr (up to 21.0 wt.% Cr_2O_3). Aluminous magnesian titanomagnetites are also found, but most magnetites in the ultramafic lamprophyres are pure magnetite. The compositional variation of ilmenite from the Sokli complex is mainly caused by the substitution of Fe^{2+} into Mg and Mn, and partly Ti into Nb. Mg- and Mn-rich ilmenites in the early stage P1-C1 rocks evolve towards pure $FeTiO_3$ composition in the latest D5 dolomite carbonatite. Ilmenites from the ultramafic lamprophyres are relatively poor in Mn compared to those from the phoscorite-carbonatite complex. In the coexisting magnetite and ilmenite, Mg and Mn cations preferentially partition into ilmenite rather than magnetite. In spite of the mineralogical and geochemical contrasts between the phoscorites and associated carbonatites, compositions of Fe-Ti oxide minerals from the two paired rocks at a given stage are basically the same, and evolve systematically from stage to stage. This supports the hypothesis that the phoscorites and conjugate carbonatites are derived from common parental melts. Furthermore, the intrusions of the Sokli phoscorite-carbonatite complex are considered to have either crystallized from successive batches representing a continuum in time and magmatic evolution, or that the P1-C1 parental magma has simply differentiated to form the later stage rocks.

Key words: magnetite, ilmenite, phoscorite, carbonatite, ultramafic lamprophyre, Sokli complex

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1. INTRODUCTION

Fe-Ti oxides are principal accessory constituents, but are locally essential phases, in carbonatites and genetically related rocks, including phoscorites (consisting essentially of forsterite, apatite, magnetite and carbonates) and various alkaline silicate rocks. Although the crystal chemistry and thermochemistry of Fe-Ti oxides are complex (e.g., Sack, 1982; Ghiorso and Sack, 1991), their compositions are sensitive indicators of intensive and extensive magmatic variables (Haggerty, 1991a; Haggerty, 1994). Moreover, they have a high capacity for cationic substitutions in Fe- and Ti-rich phoscorite-carbonatite system, and thus are important minerals for tracing the geochemical evolution of phoscorite-carbonatite complexes. Several studies have produced analyses of Fe-Ti oxide minerals from carbonatites; in East Africa (Prins, 1972), Sarfartôq, Greenland (Secher and Larsen, 1980) and Jacupiranga, Brazil (Mitchell, 1978; Gaspar and Wyllie, 1983a; Gaspar and Wyllie, 1983b). These studies show that most magnetite compositions are near to pure magnetite (less than 2 wt.% TiO_2, less than 1 wt.% Al_2O_3, MgO and MnO), but ilmenite shows a wide range from Mg- and Mn-enriched composition to almost pure $FeTiO_3$.

The Sokli complex in north eastern Finland (360 Ma, Kramm et al., 1993) is one of numerous carbonatite-alkaline complexes in the Kola Alkaline Province and bears the largest Nb mineral deposit in Europe. The complex is also a host for vast phosphorous ores. The complex comprises multi-intrusions of phoscorites and carbonatites in the center and ultramafic rocks in the outer regions. Numerous veins and dikes of ultramafic lamprophyres ranging from a few centimeters to several meters in thickness cut across all constituent members of the complex. Outcrop conditions are very poor, but intensive campaigns of drilling and trenching for exploring of economic deposits provide fresh materials for the systematic study on Fe-Ti oxide minerals of the intrusive phases. In this paper, we present chemical data for Fe-Ti oxide minerals from early to late carbonatite-phoscorite associations and ultramafic lamprophyres of the complex and compare their chemistry with those from world-wide carbonatites and ultramafic rocks. Furthermore, we explore the applications for the petrogenesis of the Sokli

rocks by tracing compositional variations of them throughout the whole complex.

2. GENERAL GEOLOGY AND PETROGRAPHY

The Sokli complex is a member of the Kola Alkaline Province which consists of more than 20 massifs of the Devonian ultramafic-alkaline rocks (Woolley, 1989; Kramm et al., 1993; Kogarko et al., 1995). Sixteen of these complexes contain carbonatites. A characteristic feature of the majority of Kola carbonatite complexes is their close association with phoscorites, rocks consisting essentially of apatite-magnetite and silicates (forsterite and/or phlogopite). Rare metal mineralizations are in close relation with the differentiation of these phoscorite-carbonatite complexes.

2.1. Phoscorite and Carbonatite Associations

The Sokli complex has a concentrically zoned structure, is divided into two zones, and is surrounded by a 1–2 km fenite aureole (Fig. 1). The outer zone consists mainly of ultramafic rocks which were largely transformed into carbonate-bearing metasomatic facies by CO_2-rich fluids derived from later carbonatite injections. The inner zone (phoscorite-carbonatite complex, PCC), or "magmatic core", is made up of multi-intrusions of carbonatites and phoscorites. In the magmatic core, five intrusive stages of phoscorites and carbonatites are distinguished based on mineral assemblages and bulk rock compositions (Lee et al., 2003a; Lee et al., 2004). The first three stages comprise phoscorites and calcite carbonatites (referred to as P1 to P3 and C1 to C3) and they usually occur as pairs with the same mineral assemblages. The last two stages comprise only dolomite carbonatites called D4 and D5.

Phoscorites and calcite carbonatites of the first three groups are coarse to medium grained rocks composed mainly of calcite, apatite, magnetite, olivine and mica. Enrichment in calcite visually distinguishes the carbonatites from the phoscorites. Conversely, the phoscorites are enriched in magnetite and silicates, as well as accessory Nb-Zr minerals (baddeleyite, pyrochlore and zirconolite), dolomite and sulfides. Significant modal variations are found within each rock type, but gradational transitions from carbonatite to phoscorite have not been observed. Modal proportions of carbonate minerals and the ratio between calcite and dolomite substantially vary from stage to stage. In the P1C1 group, the contrast between the paired rocks is relatively small (10–30 vol.% calcite in P1, 60–80 vol.% in C1); this contrast increases in the P2C2 and P3C3 groups, i.e., the calcite carbonatites become increasingly calcite-rich, and the phoscorites increasingly calcite-poor, while secondary dolomite becomes more abundant. This trend towards more leucocratic carbonatites and more dolomitic systems culminates in D4, which are very poor in silicates, magnetite and apatite. In the latest D5 group, dolomite becomes enriched in Fe and Mn and a new set of rare Sr-Ba-LREE-bearing minerals and sulfides appear, including ancylite and pyrite.

2.2. Ultramafic Lamprophyre

Ultramafic lamprophyres (UML), reviewed by Rock (1991), are made up of phenocrysts of one or more of minerals, Mg-olivine, Ti-phlogopite and Ti-augite in a matrix of carbonate, melilite, feldspathoids, monticellite, ilmenite, perovskite, Cr-spinel and Ti-magnetite. The most common types of UML are alnöite (melilite-rich, clinopyroxene-bearing) and aillikite (melilite-free, carbonate-rich).

The Sokli UML occur as dikes on the scale of millimeters
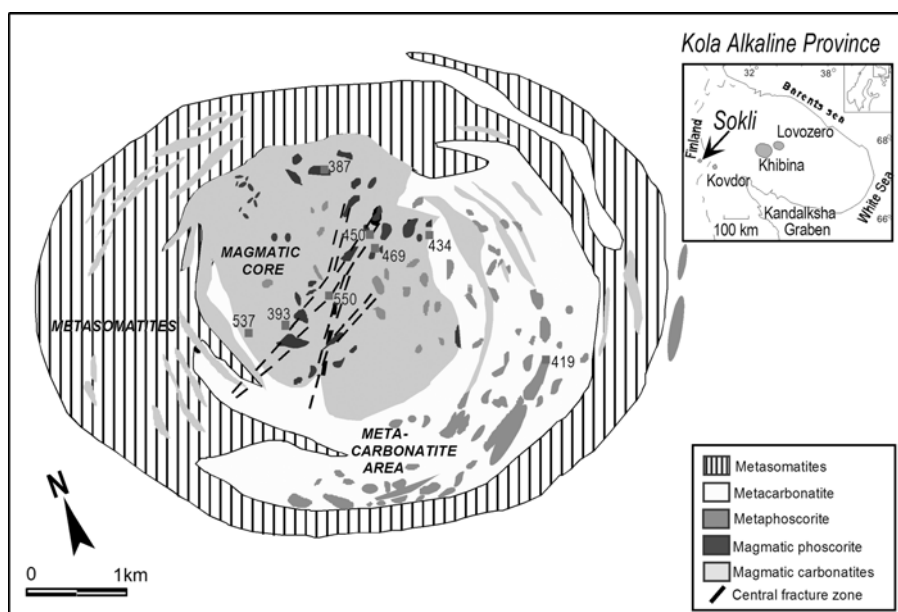

Fig. 1. Map of the Sokli complex (modified from Vartiainen, 1980) showing the location of sampling drill holes and distribution of five stage phoscorites and carbonatites. Inset: Map of the Kola Alkaline Province (after Bell et al., 1996).

to meters. They cut most rock types, but some lamprophyric dikes are assimilated and brecciated by later carbonatitic intrusions. Petrography of the Sokli UML has been described by Vartiainen et al. (1978) and Lee et al. (2003b). They have various modal proportions of forsterite and Ti-phlogopite phenocrysts and/or xenocrysts in a calcite-rich groundmass, and hence they are not considered to be in textural or chemical equilibrium. Petrographical and geochemical characteristics of the Sokli UMLs indicate that they have affinities with aillikites.

Two varieties of lamprophyres have been identified: one is a medium-grained, mica-rich variety, and the other is a porphyritic, olivine-rich one. Mica-rich lamprophyre is distinguished by high modal proportion of phlogopite (up to 60%) and the rarity of olivine phenocrysts. On the other hand, porphyritic olivine-rich variety is characterized by common macrocrysts of olivine and phlogopite. Olivines are usually more abundant than phlogopites, and they are set in a fine grained magnetite and calcite matrix. Both varieties basically share the same mineral assemblage, but their modal proportions are significantly different. The large compositional and textural variations observed are not considered to be the result of derivation from different parental magmas, but instead reflect their highly hybrid nature. This is supported by the presence of varying proportions of olivine and phlogopite xenocrysts.

2.3. Occurrence of Fe-Ti oxides

Magnetite is ubiquitously found within all rock facies of the Sokli complex. It commonly occurs as disseminated euhedral grains or as lenses and irregular aggregates. Two generations of magnetite are distinguished in the Sokli PCC. The early generation is usually large crystals (up to 2 cm) and associates with olivine or calcite. These are common in stage 1 and 2 rocks. The late generation occurs as small (tens of m) euhedral crystals and appears from stage 3 rocks. In contrast to the late generation, the early magnetite commonly contains internal or external inclusions of ilmenite.

Early magnetite commonly occurs as vermicular intergrowths with calcite or olivine in the C1 carbonatite. Discrete euhedral magnetite is rare. In the C2 carbonatite, vermicular and skeletal intergrowths are also encountered, however large euhedral magnetite crystals ranging from 2 to 4 mm in size is more common. Modal proportions of magnetite decrease toward C3 carbonatite where magnetite occurs as small euhedral grains. Magnetite is rare in the late stage D4 and D5 carbonatites. Sulfide minerals are the dominant opaques in dolomite carbonatites.

Magnetite is one of the major constituents of phoscorite. Modal percentages and grain size are extremely variable. The volume varies from 15 to 70%, the grain size ranges from 1 to 20 mm, and both variables tend to increase from P1 to P3. Occasionally, magnetite displays internal fractures

and is corroded. In P1 and P2, magnetite forms, with apatite aggregates, large coalescent clusters that enclose euhedral olivine grains. The whole structure is cemented by later phlogopite or replaced with pools of carbonate.

In the UML, magnetite occurs as phenocrysts or xenocrysts and the morphology varies from euhedral to anhedral. The anhedral variety forms aggregates of small crystals. Ilmenite usually occurs as internal or external granules in early magnetite, which has been described as composite type ilmenite intergrowths (Haggerty, 1991b). It is rarely found as a form of lamellar exsolution. Ilmenite is also present as individual crystals with poorly developed rhombohedral faces. These particular ilmenites crystallized after the early magnetite and the majority of silicates. Composite type ilmenite intergrowths are commonly observed in the stage 1 and 2 rocks (Fig. 2a, b), however, discrete ilmenite crystals are more common in the stage 3 rocks. All varieties of ilmenite, i.e., lamellae and composite type exsolutions and individual grains are found in the Sokli UML.

Two morphological types of lamellar ilmenite are observed in back-scattered electron images (BSE, Fig. 2c, d). There are long thin lamellae oriented parallel to {111} of the magnetite host, and coarse irregular laths distributed at random. These two types of lamellae can be classified as trellis and sandwich type lamellae, respectively (Haggerty, 1991b). The trellis type lamellar is found only in some UML samples (Fig. 2c) and usually coexists with the sandwich type laths. Sandwich type laths are found in stage 1 and 2 phoscorites and carbonatites and are predominatly concentrated at the margin of magnetite crystals. This suggests that the lamellae were produced by oxidation of initially homogeneous titaniferous magnetite.

3. CHEMICAL COMPOSITION

Fe-Ti oxide mineral compositions were determined using a CAMECA SX 50 wavelength-dispersive electron microprobe at the University of Paris VI and the Korea Basic Science Institute. Although standards used in the analyses are different in the two laboratories, operating conditions were the same: 15 kV accelerating voltage, 10 nA beam current, 10 μm spot size and 20 seconds counting times.

Magnetite and ilmenite analyses listed in Tables 1 and 2 have been normalized with 4 oxygens for magnetite and 3 oxygens for ilmenite. Additional elements beyond those listed in the analyses are below detection limits. Calculated Fe^{3+} values thus represent maximum contents because other elements not analyzed here occupy space which is theoretically allocated to Fe^{3+}.

3.1. Magnetite

Electron microprobe analyses reveal that magnetite of the Sokli complex belongs essentially to the magnetite-ulvöspinel
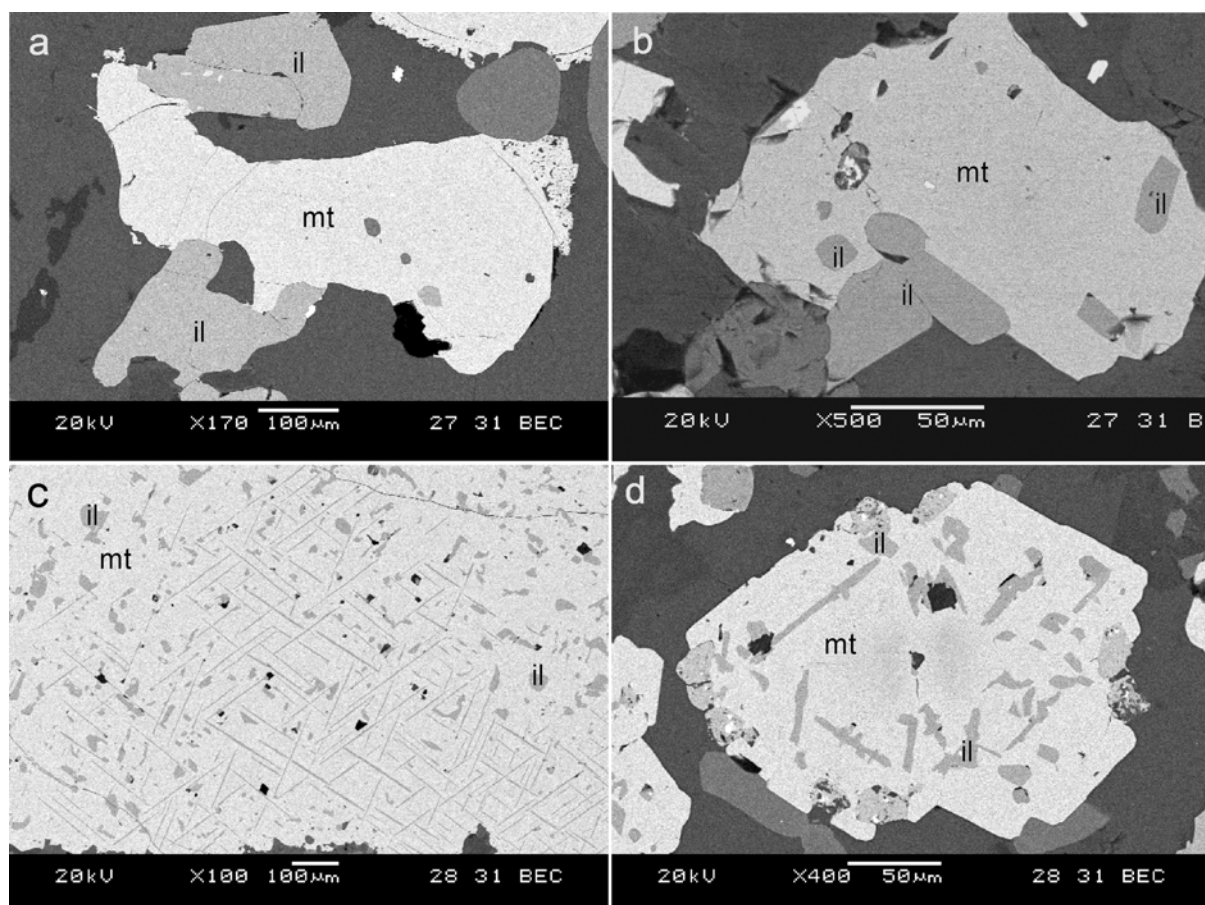
Fig. 2. Backscattered electron images of magnetite (mt) and ilmenite (il) from the Sokli complex. (a) External composite ilmenites associated with magnetite from P1; (b) internal and external composite inclusions of ilmenite in the magnetite host from C2; (c) coexisting trellis and sandwich type exsolution lamellae of ilmenite in the magnetite host from the UML; (d) sandwich type lamellae of ilmenite in the magnetite host from C2.

solid solution series with small proportions of magnesioferrite (Table 1). Compositional variations of magnetite are shown in Figure 3. Magnetite compositions from the Turiy carbonatites (Kola Alkaline Province), the Vuorijarvi carbonatites (Kola Alkaline Province) and the Jacupiranga carbonatites (Brazil) are illustrated together for comparison. $Fe^{2+}/(Fe^{2+}+Mg)$ ratio, and Al, Ti and Mg contents were used to evaluate differentiation trends. Al and Mg contents have broadly positive correlation and are the most important valuables in distinguishing magnetite from different stage rocks at Sokli (Fig. 3).

Magnetites from other carbonatites show generally similar compositional ranges with those from the Sokli varieties, but the Jacupiranga magnetite has a considerably different composition, displaying higher Mg and lower Ti and Mn contents. Magnetites from the Kola carbonatites have higher $Fe^{2+}/(Fe^{2+}+Mg)$ ratios and lower $Fe^{3+}/(Fe^{2+}+Fe^{3+})$ ratios compared with those from the Jacupiranga carbonatites. However, the total FeO content of most magnetites illustrated in Figure 3 remains in a narrow band, around 85

wt.%, and most of them show an inverse correlation between $Fe^{3+}/(Fe^{2+}+Fe^{3+})$ and Ti (Fig. 3c). This implies that the dominant substitution in carbonatites from the Kola Alkaline Province and Jacupiranga was made within the magnetite-ulvospinel series.

Magnetite from the Sokli PCC shows a systematic compositional variation from stage 1 to the final stage 5 with increasing $Fe^{2+}/(Fe^{2+}+Mg)$ ratio, and decreasing in Ti, Al and Mn contents. The early generation of magnetite from stage 1 and 2 rocks is relatively far from the pure $Fe^{3+}_2Fe^{2+}O_4$ composition: Fe^{2+} is largely substituted by Mg^{2+} and Mn^{2+}, and Fe^{3+} by Ti^{4+} combined with Fe^{2+}. Magnetite from the P1C1 rocks is distinguished by its high Mg, Al and Mn contents (Fig. 3). Compared to the early stage magnetite, the late generation from stage 3, 4 and 5 rocks is significantly poor in Mg, Mn, Al and Ti and thus has near pure magnetite composition. The absence of ilmenite inclusions in the late stage magnetite and the small grain sizes suggest rapid crystallization, probably combined with very low $f(O_2)$ system. It is noteworthy that compositions of magnetite from

Table 1. Representative chemical compositions of magnetites.

Sample	434R219	434R219	434R203	434R203	SS4	SS4	528R244	528R244	434R251	434R251	393R150	393R150	393R150	393R150	393R174	393R174	480R80	480R80
Rock-type	C1	C1	P1	P1	C2	C2	P2	P2	C3	C3	P3	P3	P3	P3	P3	P3	D4	D4
SiO_2	0.06	0.08	0.19	0.16	0.06	0.08	0.07	0.07	0.07	0.06	0.18	0.14	0.11	0.32	0.26	0.20	0.09	0.05
TiO_2	1.72	5.54	4.17	4.62	1.42	1.08	2.18	0.82	0.80	0.66	1.33	1.67	1.47	2.19	1.90	12.78	0.64	0.56
Nb_2O_5	-	-	0.04	0.02	-	-	0.04	-	0.05	0.05	0.04	-	-	0.03	-	-	-	0.22
Al_2O_3	0.69	0.62	1.32	1.50	0.07	0.09	0.05	0.04	0.08	0.04	0.04	0.02	0.02	0.04	0.02	-	0.05	-
Cr_2O_3	-	0.01	0.02	0.03	-	0.03	0.03	0.02	0.02	0.03	0.04	0.02	n.d.	0.02	0.04	0.02	0.05	0.06
V_2O_3	n.a.	n.a.	n.a.	n.a.	-	-	-	-	-	-	0.28	0.30	0.38	0.22	0.22	n.a.	n.a.	n.a.
Fe_2O_3	66.08	59.01	61.44	60.52	66.74	67.82	66.08	67.35	67.95	68.40	65.71	64.82	65.58	64.13	64.52	45.71	67.42	67.57
FeO	27.74	29.70	28.07	27.72	27.94	28.68	29.09	29.47	28.95	29.49	31.82	31.56	31.13	32.86	31.75	35.74	30.76	31.18
MgO	2.56	3.32	3.87	4.27	2.28	1.97	2.24	1.13	1.48	1.30	0.60	0.85	1.18	0.68	0.84	3.49	0.37	0.33
MnO	1.06	1.48	1.53	1.67	0.76	0.48	0.79	0.45	0.63	0.36	0.42	0.37	0.45	0.23	0.55	2.05	0.19	0.13
CaO	-	-	0.02	-	-	0.03	0.04	0.03	-	0.01	0.07	-	-	-	-	-	0.07	0.07
NiO	0.08	-	0.09	-	-	-	-	-	0.06	0.06	0.04	0.07	-	0.04	-	-	0.04	n.d.
Total	99.99	99.75	100.75	100.50	99.27	100.25	100.59	99.39	99.97	100.44	100.56	99.79	100.31	100.70	100.09	99.98	99.62	100.18
Number of cations on the basis of 4 Oxygens																		
Si	0.002	0.003	0.007	0.006	0.002	0.003	0.002	0.003	0.003	0.002	0.007	0.005	0.004	0.012	0.010	0.007	0.004	0.002
Ti	0.049	0.155	0.115	0.127	0.041	0.031	0.062	0.024	0.023	0.019	0.038	0.048	0.042	0.062	0.055	0.356	0.019	0.016
Nb	-	-	0.001	-	-	-	0.001	-	-	0.001	0.001	-	-	-	-	-	-	0.004
Al	0.031	0.027	0.057	0.065	0.003	0.004	0.002	0.002	0.004	0.002	0.001	-	-	0.001	-	-	0.001	-
Cr	-	-	-	0.001	-	0.001	0.001	0.001	-	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.002
V	-	-	-	-	-	-	-	-	-	-	0.009	0.009	0.011	0.007	0.007	0.001	-	-
Fe^{3+}	1.868	1.656	1.696	1.667	1.911	1.928	1.867	1.945	1.945	1.953	1.880	1.865	1.873	1.829	1.850	1.273	1.954	1.950
Fe^{2+}	0.871	0.927	0.861	0.849	0.889	0.906	0.913	0.946	0.921	0.936	1.012	1.009	0.988	1.042	1.012	1.106	0.991	1.000
Mg	0.143	0.185	0.211	0.233	0.130	0.111	0.126	0.065	0.084	0.073	0.034	0.048	0.067	0.038	0.048	0.193	0.021	0.019
Mn	0.034	0.047	0.048	0.052	0.025	0.015	0.025	0.015	0.020	0.012	0.014	0.012	0.015	0.007	0.018	0.064	0.006	0.004
Ca	-	-	0.001	-	-	0.001	0.002	0.001	-	0.001	0.003	-	-	-	-	-	0.003	0.003
Ni	0.003	-	0.003	-	-	-	-	-	0.002	0.002	0.001	0.002	-	0.001	-	-	0.001	-

FeO and Fe_2O_3 are calculated by Droop's (1987) method. n.a., not analyzed; -, below detection limit. See text for the definition of rock types

Table 1. (continued)

Sample	537R166	537R166	419R87	419R87	419R87	434R185	434R185	469R173	469R173	469R173	450R196	450R196	450R196	450R196	450R196	450R196	450R196
Rock-type	D5	D5	UML	UML	UML	UML	UML	UML	UML	UML	UML	UML	UML	UML	UML	UML	UML
SiO_2	0.24	0.17	0.05	0.05	0.04	0.17	0.16	0.27	0.16	0.15	0.09	0.08	0.10	0.09	0.10	1.75	0.08
TiO_2	1.12	1.64	11.26	10.93	11.02	3.07	2.81	1.51	1.10	1.80	2.73	3.16	1.00	3.65	4.04	1.88	1.63
Nb_2O_5	0.08	-	-	-	0.03	-	-	-	0.10	0.07	-	-	-	-	-	0.11	-
Al_2O_3	0.19	0.52	1.46	1.46	1.55	0.01	-	0.07	0.10	0.07	-	-	-	-	-	-	-
Cr_2O_3	0.02	-	0.02	0.03	-	1.02	0.07	0.05	0.04	0.03	10.60	20.95	5.25	9.83	10.86	1.50	0.59
V_2O_3	0.08	0.10	n.a.	n.a.	n.a.	0.39	0.28	0.51	0.48	0.40	0.47	0.22	0.40	0.26	0.26	0.26	0.28
Fe_2O_3	65.53	65.13	47.11	47.62	47.23	61.62	63.09	64.54	65.10	64.15	51.61	41.66	60.60	51.92	49.84	59.79	64.93
FeO	29.85	30.00	34.00	33.12	33.49	32.15	32.40	30.64	29.88	29.83	30.60	29.09	30.24	30.49	30.25	31.31	31.23
MgO	1.33	1.60	4.45	4.64	4.49	1.52	1.13	1.53	1.52	1.66	1.76	2.46	1.06	2.14	2.33	2.46	0.95
MnO	0.38	0.48	0.68	0.81	0.75	0.36	0.25	0.84	0.75	1.02	1.19	1.72	0.43	1.27	1.38	0.34	0.35
CaO	-	0.02	-	-	-	0.03	0.02	-	-	-	-	-	0.40	-	-	0.02	0.18
NiO	0.02	-	-	0.04	0.08	0.20	0.08	-	-	0.06	-	0.05	0.03	0.06	0.15	0.17	0.07
Total	98.82	99.64	99.04	98.69	98.67	100.55	100.28	99.98	99.15	99.17	99.04	99.43	99.51	99.72	99.20	99.59	100.28
Number of cations on the basis of 4 Oxygens																	
Si	0.009	0.006	0.002	0.002	0.002	0.007	0.006	0.010	0.006	0.006	0.003	0.003	0.004	0.003	0.004	0.066	0.003
Ti	0.032	0.047	0.312	0.304	0.307	0.087	0.080	0.043	0.032	0.052	0.078	0.089	0.029	0.103	0.115	0.053	0.047
Nb	0.001	-	-	-	-	-	-	-	-	-	-	-	-	-	-	0.002	-
Al	0.008	0.023	0.064	0.063	0.067	-	-	0.003	0.005	0.003	-	-	-	-	-	0.000	-
Cr	-	-	-	0.001	-	0.030	0.002	0.001	0.001	0.001	0.318	0.621	0.158	0.292	0.324	0.045	0.018
V	0.002	0.003	-	-	-	0.012	0.008	0.016	0.015	0.012	0.014	0.007	0.012	0.008	0.008	0.008	0.009
Fe^{++}	1.897	1.861	1.307	1.324	1.315	1.747	1.801	1.841	1.874	1.844	1.476	1.175	1.740	1.471	1.416	1.689	1.857
Fe^{2+}	0.960	0.953	1.049	1.024	1.036	1.013	1.028	0.971	0.956	0.953	0.972	0.912	0.965	0.960	0.955	0.983	0.993
Mg	0.076	0.090	0.244	0.256	0.247	0.085	0.064	0.087	0.087	0.094	0.100	0.138	0.060	0.120	0.131	0.138	0.054
Mn	0.012	0.015	0.021	0.025	0.023	0.011	0.008	0.027	0.024	0.033	0.038	0.055	0.014	0.040	0.044	0.011	0.011
Ca	-	0.001	-	-	-	0.001	0.001	-	-	-	-	-	0.017	-	-	0.001	0.007
Ni	0.001	-	-	0.001	0.002	0.006	0.002	-	-	0.002	-	0.001	0.001	0.002	0.004	0.005	0.002

Table 2. Representative chemical compositions of Ilmenites.

Sample	434R219	434R219	434R201	434R201	434R201	SS4	SS4	SS4	528R244	528R244	528R244	450R157	450R157	393R223	393R223	393R150	393R150
Rock-type	C1	C1	P1	P1	P1	C2	C2	C2	P2	P2	P2	C3	C3	C3	C3	P3	P3
SiO_2	0.05	0.04	0.04	0.06	0.03	0.08	0.08	0.14	0.09	0.08	0.06	0.03	0.05	0.06	0.05	0.21	0.13
TiO_2	59.21	59.13	58.81	56.44	56.61	57.45	57.50	49.55	53.03	55.57	54.79	50.64	51.92	50.06	50.57	53.10	51.95
Nb_2O_5	0.06	-	0.81	0.55	0.30	0.37	0.47	0.61	2.04	0.37	0.35	1.62	0.89	1.61	1.46	0.82	0.96
Cr_2O_3	0.01	-	0.01	-	-	-	0.02	-	0.01	0.03	0.03	-	0.04	0.03	0.02	0.03	-
Fe_2O_3	-	-	-	-	1.14	-	-	9.69	2.29	1.00	0.63	4.00	3.49	2.42	2.03	2.83	2.51
FeO	18.51	16.53	17.71	24.58	23.93	20.07	20.16	19.44	27.85	27.91	31.35	33.16	34.06	43.08	43.06	33.63	37.04
MgO	14.76	13.21	14.95	10.55	11.18	11.37	11.56	9.28	9.28	9.63	6.66	5.14	5.11	0.89	1.13	5.95	3.20
MnO	7.65	11.96	8.59	7.62	7.22	10.83	10.88	9.19	5.49	5.30	6.39	4.87	4.38	2.06	1.99	4.51	5.06
CaO	n.d.	0.05	0.01	0.05	0.05	0.01	0.03	0.06	0.03	0.02	0.01	0.03	0.06	0.05	0.02	-	0.04
NiO	0.02	0.01	-	0.01	0.02	-	-	0.02	-	-	-	0.04	0.02	-	0.01	0.08	0.04
Total	100.26	100.93	100.93	99.87	100.49	100.18	100.69	97.97	100.10	99.89	100.26	99.52	100.00	100.28	100.32	101.16	100.89
Number of cations on the basis of 3 Oxygens																	
Si	0.001	0.001	0.001	0.001	0.001	0.002	0.002	0.003	0.002	0.002	0.001	0.001	0.001	0.002	0.001	0.005	0.003
Ti	1.005	1.008	0.994	0.992	0.985	1.001	0.995	0.899	0.945	0.983	0.987	0.935	0.952	0.948	0.955	0.956	0.958
Nb	0.001	-	0.008	0.006	0.003	0.004	0.005	0.007	0.022	0.004	0.004	0.018	0.010	0.018	0.017	0.009	0.011
Cr	-	-	-	-	-	-	-	-	-	-	0.001	-	0.001	0.001	-	-	-
Fe^{3+}	-	-	-	-	0.020	-	-	0.176	0.041	0.018	0.011	0.074	0.064	0.046	0.038	0.051	0.046
Fe^{2+}	0.350	0.313	0.333	0.481	0.463	0.389	0.388	0.392	0.552	0.549	0.628	0.681	0.694	0.907	0.904	0.673	0.759
Mg	0.497	0.447	0.501	0.368	0.385	0.392	0.397	0.334	0.328	0.338	0.238	0.188	0.186	0.034	0.042	0.212	0.117
Mn	0.146	0.230	0.164	0.151	0.142	0.212	0.212	0.188	0.110	0.106	0.130	0.101	0.091	0.044	0.042	0.092	0.105
Ca	-	0.001	-	0.001	0.001	-	0.001	0.001	0.001	-	-	0.001	0.001	0.001	-	-	-
Ni	-	-	-	0.001	0.001	-	0.001	0.001	0.001	-	-	0.001	0.001	-	-	0.002	0.001

FeO and Fe2O3 are calculated by Droop's (1987) method. -, below detection limit. See text for the definition of rock types

Table 2. (continued)

Sample	393R150	393R174	393R174	550R196	550R196	387R71	387R71	434R185	434R185	434R185	434R185	434R185	434R185	450R196	469R173
Rock-type	P3	P3	P3	D5	D5	UML	UML	UML	UML	UML	UML	UML	UML	UML	UML
SiO_2	0.09	0.19	0.15	0.13	0.26	0.07	0.15	0.20	0.11	0.18	0.15	0.16	0.09	0.10	0.53
TiO_2	52.27	52.72	52.59	49.88	49.26	51.74	51.84	44.87	50.30	54.13	54.87	54.38	53.92	53.70	53.39
Nb_2O_5	0.91	0.83	0.92	1.34	1.88	2.51	2.25	-	0.10	0.02	0.10	0.11	0.34	0.19	0.48
Cr_2O_3	0.01	-	0.02	-	-	0.11	0.06	3.67	0.81	0.06	0.08	0.05	0.08	-	-
Fe_2O_3	2.61	2.36	2.98	1.64	0.66	2.24	2.54	17.03	9.93	3.67	2.39	4.96	3.84	2.35	1.79
FeO	36.99	34.71	34.79	40.57	42.73	29.99	30.20	26.12	29.83	32.31	32.74	30.33	33.11	35.07	24.91
MgO	3.41	4.81	4.82	1.19	0.41	8.12	7.94	7.24	7.87	8.53	8.63	9.67	8.02	5.63	5.41
MnO	4.95	5.15	5.00	3.72	3.04	4.78	4.74	1.42	1.50	1.39	1.47	1.61	1.46	3.37	14.18
CaO	-	-	-	-	0.04	0.02	0.02	-	0.03	-	-	-	-	0.03	0.14
NiO	-	0.03	-	-	0.05	0.00	0.03	0.12	0.05	-	0.02	-	-	0.06	0.07
Total	101.23	100.80	101.27	98.47	98.33	99.58	99.78	100.66	100.51	100.29	100.45	101.26	100.93	100.49	100.88
Number of cations on the basis of 3 Oxygens															
Si	0.002	0.005	0.003	0.003	0.007	0.002	0.004	0.005	0.003	0.004	0.003	0.004	0.002	0.002	0.013
Ti	0.959	0.960	0.954	0.958	0.954	0.936	0.936	0.807	0.899	0.962	0.973	0.951	0.958	0.973	0.963
Nb	0.010	0.009	0.010	0.015	0.022	0.027	0.024	-	-	-	0.001	0.001	0.004	0.002	0.005
Cr	-	-	-	-	-	0.002	0.001	0.069	0.015	-	0.001	0.001	0.001	-	-
Fe^{3+}	0.048	0.043	0.054	0.031	0.013	0.040	0.046	0.307	0.178	0.065	0.042	0.087	0.068	0.043	0.032
Fe^{2+}	0.755	0.703	0.702	0.866	0.920	0.603	0.607	0.522	0.593	0.638	0.645	0.590	0.654	0.707	0.500
Mg	0.124	0.174	0.173	0.045	0.016	0.291	0.284	0.258	0.279	0.300	0.303	0.335	0.282	0.202	0.194
Mn	0.102	0.106	0.102	0.080	0.066	0.097	0.096	0.029	0.030	0.028	0.029	0.032	0.029	0.069	0.288
Ca	-	-	-	-	0.001	-	0.001	-	0.001	-	-	-	0.002	0.001	0.004
Ni	-	0.001	-	-	0.001	-	0.001	0.002	0.001	-	-	-	-	0.001	0.001
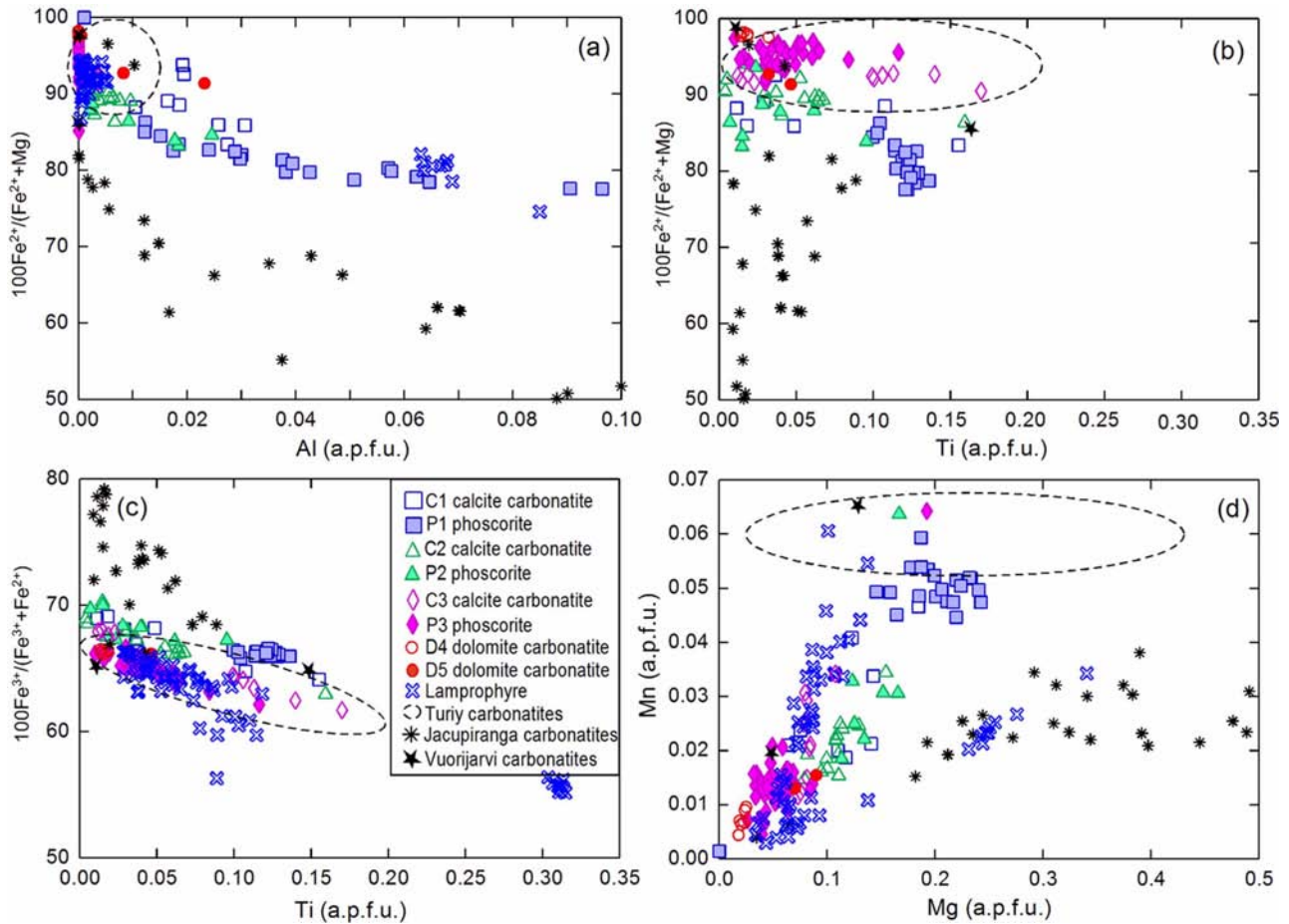
Fig. 3. Compositional variation of magnetite from the Sokli rocks. Magnetite compositions in carbonatites from Turiy (Dunworth and Bell, 2003), Vuorijarvi (Stephane, 2001) and Jacupiranga (Gaspar and Wyllie, 1983a) are plotted together.

phoscorites and the paired carbonatites within the same stage are broadly similar like as phlogopite compositions of them (Lee et al., 2003a).

It is known that magnetite from plutonic carbonatites is normally very poor in Ti, but typically richer in Mg and Al (Kukharenko et al., 1965; Prins, 1972; Secher and Larsen, 1980; Gapar and Wyllie, 1983a). However, the Ti content of magnetite in carbonatites of the Kola alkaline Province, including the Turiy and Vuorijarvi carbonatites, which are associated with phoscorites, is relatively higher than those of other world-wide carbonatite magnetites (Fig. 3). Even if several magnetites analyzed from late stage rocks at Sokli have comparable Ti contents to those of early stage rocks. This characteristic is considered to be related to the parental phoscorite-carbonatite magma composition.

Three groups of magnetites are identified in the UML (Fig. 4). The most primitive magnetites, in terms of Fe-Ti-Cr content, occur as small anhedral aggregates and have a high amount of Cr (up to 21.0 wt.% of Cr_2O_3), with extremely low Al_2O_3 content (< 0.1 wt.%). These magnetites are chromian titanomagnetite with Fe/(Fe+Mg) ratio = 0.87–0.96, Cr/(Cr+Al) = 0.9–1.0 and Ti/(Ti+Cr+Al) = 0.27–0.60. Inter-

mediate small euhedral magnetites are richer in TiO_2 (about 11 wt.%), MgO (up to 6.2 wt.%) and Al_2O_3 (up to 2.0 wt.%) compared to other populations and are classified as aluminous magnesian titanomagnetite with Fe/(Fe+Mg) ratio = 0.74–0.82, Cr/(Cr+Al) = <0.02 and Ti/(Ti+Cr+Al) = 0.81–0.83. The most evolved varieties, which is the most common, are almost pure magnetite with Fe/(Fe+Mg) ratio = 0.91–0.96 and less than 1 wt.% of Cr_2O_3 and Al_2O_3. The second group of aluminous magnesian titanomagnetites have similar $Fe^{2+}/(Fe^{2+}+Mg)$ ratios to those of the P1C1 magnetites, but slightly higher Al_2O_3 content. Large diversity of magnetite compositions found in lamprophyres (Fig. 4, Table 1) is not considered to be the result of different parental melts, but instead suggests the presence of several generations of magnetite, including a large amount of xenocrysts

3.2. Ilmenite

The Sokli ilmenites demonstrate significant compositional variation with respect to Mg, Mn, Fe and Ti (Table 2) and are relatively poor in Fe^{3+}. The $FeTiO_3$ (ilmenite)-$MgTiO_3$ (geikeilite)-$MnTiO_3$ (pyrophanite) diagram is used to illus-
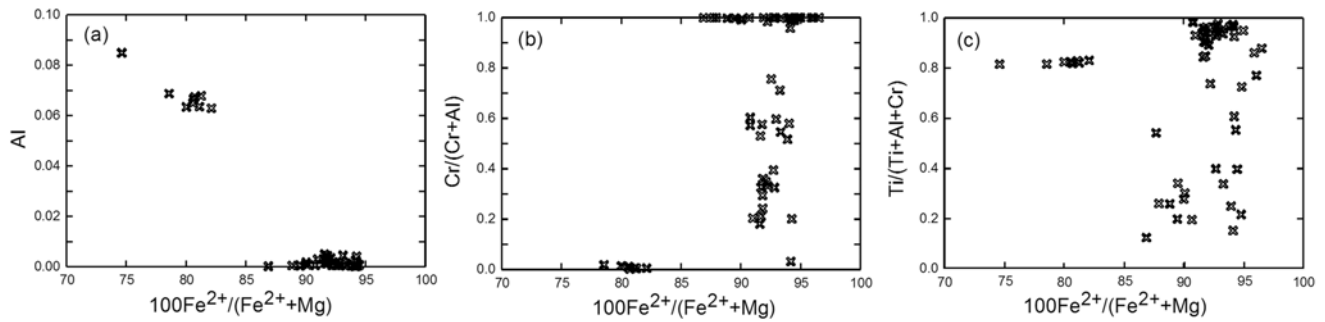
Fig. 4. Magnetite compositions (a.p.f.u.) from the Sokli UML. (a) Al, (b) Cr/(Cr+Al) and (c) Ti/(Ti+Al+Cr) vs $100Fe^{2+}/(Fe^{2+}+Mg)$.
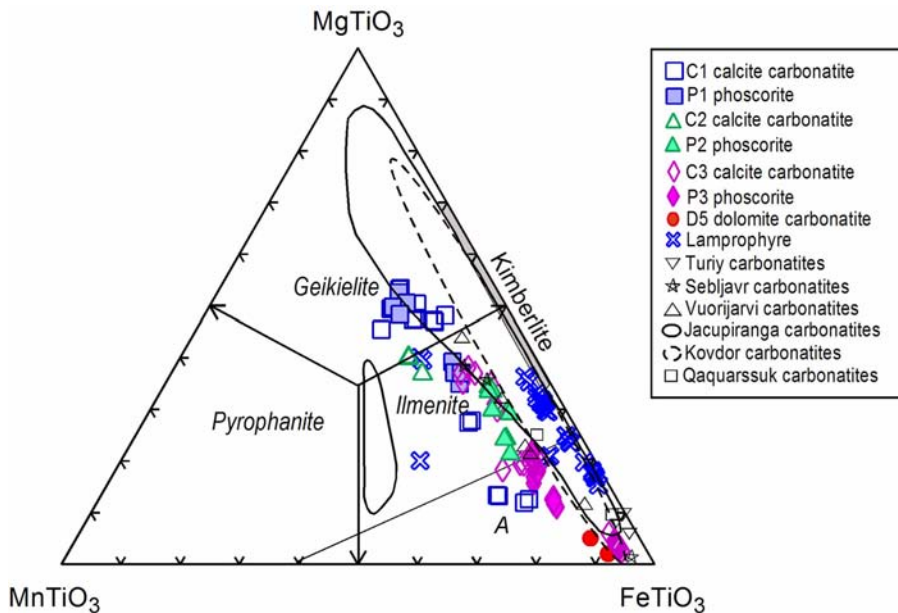
Fig. 5. Compositions of the Sokli ilmenites expressed as ternary molecular percentages of $MnTiO_3$–$MgTiO_3$–$FeTiO_3$. Field of kimberlite ilmenites from Mitchell (1973, 1977) and Haggerty (1975). Field A is for ilmenites from lamprophyres, granites, basalts and carbonatites (Bergstol, 1972; Czamanske and Mihalik, 1972; Prins, 1972; Haggerty, 1976b). Ilmenites from other world wide carbonatites are plotted together for comparison. Data are from the following references: Turiy, Kola (Garanin et al., 1980; Dunworth and Bell, 2003), Sebljavr, Kola (Garanin et al., 1980; Mitchell and Chakhmouradian, 1998), Vuorijarvi, Kola (Garanin et al., 1980), Jacupiranga, Brazil (Gaspar and Wyllie, 1983b), Kovdor, Kola (Krasnova and Krezer, 1995) and Qaquarssuk, Greenland (Chakhmouradian and Zaitsev, 1999).

trate the compositional variation of ilmenite (Fig. 5). Most ilmenites in the Sokli complex show a continuous variation from geikielite to ilmenite.

Early ilmenites from the Sokli phoscorite-carbonatite pairs occur as sandwich-type lamellae in magnetite and generally have high MgO (up to 11.4 wt.%) and MnO (up to 14.2 wt.%) contents (Table 2 and Fig. 5). This is much higher than those of ilmenites from other carbonatites and alkaline rocks (Fig. 5). Ilmenite of the Kovdor carbonatites (Krasnova and Krezer, 1995) is relatively enriched in Mg, comparable to that of kimberlite ilmenite, but poorer in Mn compared to that of the Sokli PCC. Mg- and Mn-rich ilmenite has been previously described from carbonatites of the Jacupiranga complex, Brazil (Mitchell 1978; Gaspar and Wyllie, 1983b, Fig. 5). In contrast to Sokli, the most Mn-rich ilmenite at Jacupiranga shows an extreme enrichment in Mn and occurs as discrete crystals, but not as inclusions in magnetite.

Mg- and Mn-rich populations in early stages (P1C1 through P2C2) evolve toward pure $FeTiO_3$ composition in the latest D5, at a constant calculated Fe_2O_3 component

with only a slight decrease in $MnTiO_3$ component (Fig. 5). Nb (substituting for Ti) content in ilmenite from the Sokli PCC remains low until the stage 2 phoscorite-carbonatite pairs (less than 1 wt.%). From stage 3 rocks slightly increases. The D5 ilmenite has the higest Nb content (up to 1.9 wt.% Nb_2O_3, Table 2).

In the UML, ilmenite, like other minerals from these rocks, shows a wide compositional variation. Compared with ilmenite from kimberlites, ilmenite in the Sokli UML has a slightly lower MgO content and has a comparably low pyrophanite molecule (Fig. 5). Some analyses from central parts of individual ilmenite grains in the UML are relatively high in Cr (up to 3.7 wt.% Cr_2O_3, Table 2). These crystals are considered to have not crystallized from the UML's parental melt, but to be xenocrystic nuclei enriched in Cr.

4. DISCUSSION

4.1. Magnetite-Ilmenite Relations

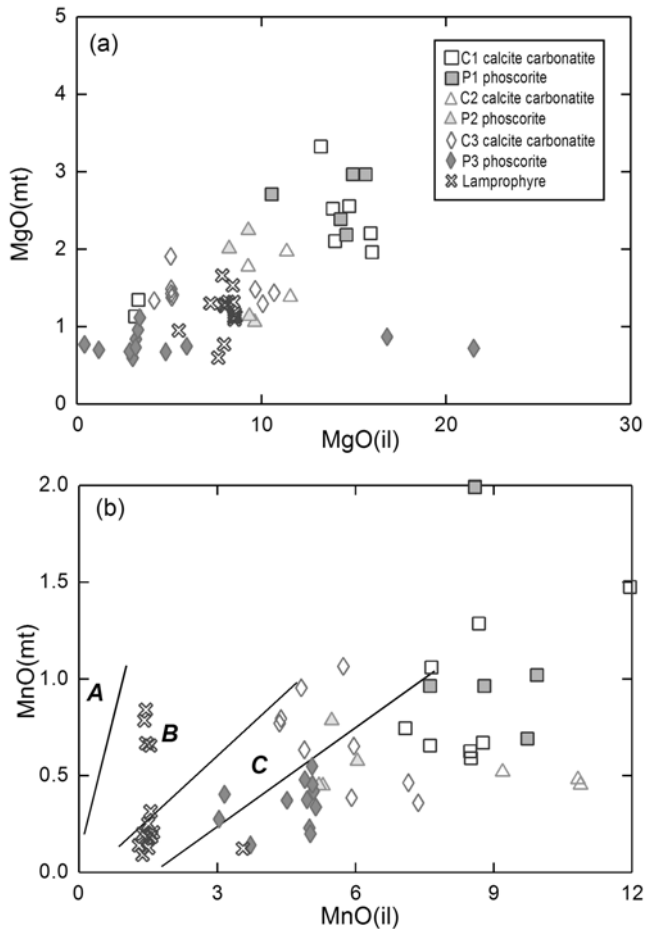The most common occurrence of ilmenite is as composite

Fig. 6. Distribution of (a) MgO and (b) MnO between coexisting magnetite and ilmenite in the Sokli rocks. The areas defined by Haggerty (1976b) are: A, basalts; B, rhyolites; C, granites; D, granodiorites, monzonites, pegmatites and syenites.

grains or sandwich type exsolutions in magnetite. These ilmenites are interpreted either as primary crystals or as products of oxidation exsolution (Haggerty, 1976a). Figure 6 compares the MgO and MnO contents of ilmenite and coexisting magnetite in the same samples. MgO and MnO contents from coexisting minerals largely show a positive correlation. The distribution of MgO and MnO between ilmenite and coexisting magnetite clearly shows a preferential incorporation of MgO and MnO into ilmenite. The preference of MnO for the ilmenite structure has been reviewed by Haggerty (1976b) in an extensive work on the crystal chemistry of Mn substitution in magnetite and ilmenite. Haggerty concluded that Mn is always strongly partitioned into ilmenite, regardless of whether ilmenite is primary or the product of oxidation exsolution, and defined the generalized area of MnO distribution as a function of rock type. The Sokli Fe-Ti oxide minerals are scattered in the area of intrusive and extrusive acidic rocks (Fig. 6).

4.2. Magmatic Evolution of the Sokli PCC

It is shown that magnetite and ilmenite from the Sokli PCC systematically change in composition from stage to stage. The general schemes are that early magnetite and ilmenite have high Mg and Mn contents; these elements gradually decrease towards late stage populations. It is also noted that the Al content of magnetite suddenly decreases from stage 2 rocks, and that late stage ilmenite contains a significant amount of Nb. These evolutionary trends of the Fe-Ti oxide minerals mainly result from Mg, Mn and Al depletions and Nb accumulation in the parental melt with time. The Ti content of magnetites from the Sokli PCC, except for some late stage magnetites, is relatively higher than those of other world-wide carbonatite magnetites. This relatively high level of Ti is presumably connected with a special characteristic of the parental phoscorite-carbonatite magma chemistry. Parental melts of PCCs are considered to bear a considerable amount of Fe, P and Si, and probably Ti, in addition to the Ca-Mg carbonate components. This assumption is supported by their mineral assemblages enriched in magnetite, apatite and fosterite and/or phlogopite (Krasnova et al., 2004; Lee et al., 2004).

All analyzed magnetites and ilmenites from the Sokli PCC show an increasing $Fe^{2+}/(Fe^{2+}+Mg)$ ratio towards the late stage populations. Therefore, we assume that the $Fe^{2+}/(Fe^{2+}+Mg)$ ratio of magnetites and ilmenites in a given stage correlates with a particular temperature and time in their crystallization history. Additionally, compositional ranges of magnetites and ilmenites from the five stage intrusions at the Sokli PCC succeed each other closely, with slight overlap (Figs. 3 and 4). This supports the view that these intrusions represent a continuum in time and chemical evolution of carbonatite magma.

5. CONCLUSIONS

The amount of magnetite in the carbonatites and phoscorites gradually increases from stage 1 towards stage 3, and crystal morphology changes from anhedral to euhedral. Magnetites from the Sokli PCC show a systematic compositional variation from stage 1 to stage 5 with increasing $Fe^{2+}/(Fe^{2+}+Mg)$ ratios, and decreasing TiO_2, Al_2O_3 and MnO contents.

The main evolution of magnetites in the Sokli phoscorite-carbonatite system is the decrease in Mg, Al, Mn and Ti contents from stage to stage, leading to pure magnetite composition in stage 5. This results mainly from the depletion of these elements in the parental melt.

The great diversity of compositions of magnetites of the UML is considered to be correlated with a large amount of xenocrysts derived from ascending lamprophyric magma from a mantle source region. Certain varieties show a high amount of Cr (up to 21.0 wt.% of Cr_2O_3), with extremely low Al_2O_3 (<0.1 wt.%). The Cr content of ilmenites in the

UML is also high compared with those of ilmenites from the phoscorite and carbonatite intrusions.

The most striking feature of the Sokli ilmenites is their unusually high MgO (up to 16 wt.%) and MnO (up to 12 wt.%) contents compared with those of ilmenites from other carbonatites. The compositional variation of the Sokli ilmenites is produced by the substitution of mainly Fe^{2+} into Mg and Mn, and partly Ti into Nb. Mg- and Mn-rich ilmenites in early stages evolve toward pure $FeTiO_3$ composition in the latest D5, with a slight decrease in Mn and increase in Nb. Mg and Mn cations are preferentially partitioned into the ilmenites rather than the coexisting magnetites in the Sokli complex.

The fact that the systematic compositional variation of magnetite and ilmenite throughout the whole phoscorite-carbonatite associations and that the basically same composition of them from the two paired rocks suggests an important key to the petrogenesis of the Sokli PCC. This supports the idea of a comagmatic origin for the phoscorites and conjugate carbonatites. As well, the Sokli PCC intrusions may represent successive rising batches of a single evolving body of magma, or that the P1C1 parental magma has simply differentiated to form the later stage rocks.

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