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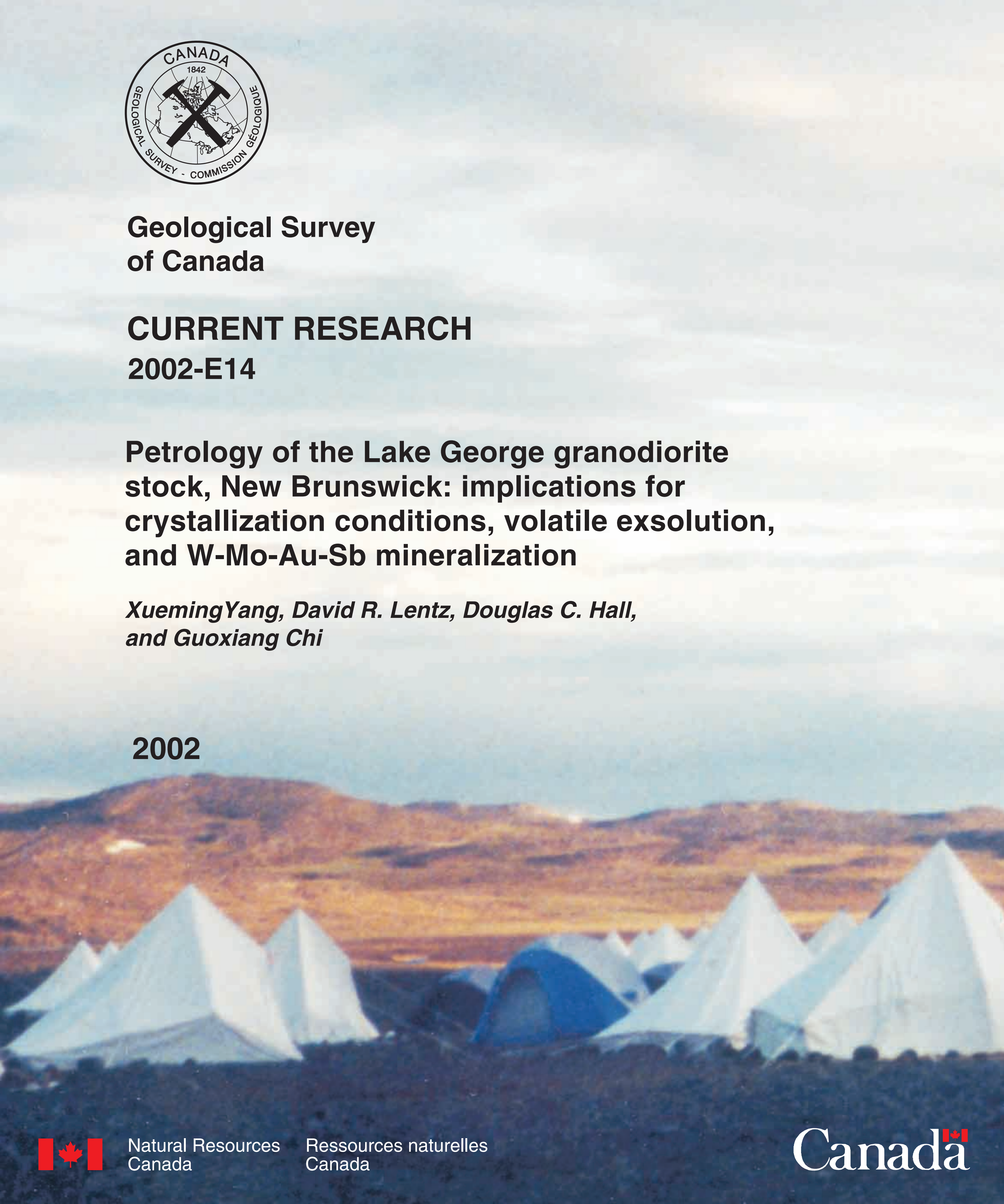

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Petrology of the Lake George granodiorite stock, New Brunswick: implications for crystallization conditions, volatile exsolution, and W-Mo-Au-Sb mineralization[1]

XuemingYang, David R. Lentz, Douglas C. Hall, and Guoxiang Chi

Yang, X., Lentz, D.R., Hall, D.C., and Chi, G, 2002: Petrology of the Lake George granodiorite stock, New Brunswick: implications for crystallization conditions, volatile exsolution, and W-Mo-Au-Sb mineralization; Geological Survey of Canada, Current Research 2002-E14, 12 p.

Abstract: The Lake George Sb vein deposit is spatially and temporally associated with W-Mo-Au mineralization that is genetically related to an Early Devonian granodiorite stock at depth. The fine- to medium-grained porphyritic granodiorite consists of quartz, plagioclase, orthoclase, biotite, and trace hornblende, with minor titanite, zircon, apatite, ilmenite, and trace magnetite. Apatite started to crystallize at 881°C to 911°C, followed by hornblende, biotite, and plagioclase at 729°C to 772°C, then quartz and orthoclase. The magmatic system was reset by late-stage subsolidus processes under 500°C. Early hornblende phenocrysts started to crystallize at 5 kbar during magma ascent, but final emplacement of the stock was epizonal (less than 2 kbar). Magmatic fluids equilibrated with biotite are characterized by Cl-rich compositions, large variations in halogen fugacity ratios, and reducing conditions; the evolving fluids emanated from the progressively crystallizing magma at depth, favouring Au partitioning from magma to the hydrothermal system.

Résumé : Le gisement filonien Lake George (tungstène) est associé dans l'espace et le temps à une minéralisation de W-Mo-Au dont l'origine est elle-même associée à un stock de granodiorite profond du Dévonien précoce. La granodiorite porphyrique à grain fin à moyen se compose de quartz, de plagioclase, d'orthose, de biotite et de traces de hornblende, ainsi que d'une quantité mineure de titanite, de zircon, d'apatite, d'ilménite et de traces de magnétite. L'apatite a commencé à cristalliser à une température comprise entre 881°C et 911 °C, suivie de la hornblende, de la biotite et du plagioclase à une température variant entre 729 °C et 772 °C, puis du quartz et de l'orthose. Des processus subsolidus tardifs ont réamorcé le système magmatique à une température inférieure à 500 °C. Les premiers phénocristaux de hornblende ont commencé à se cristalliser à 5 kbar pendant l'ascension du magma, mais la mise en place définitive du stock a été épizonale (moins de 2 kbar). Les fluides magmatiques équilibrés avec la biotite se caractérisent par de fortes teneurs en Cl, des rapports de fugacité des halogènes très variables et des conditions réductrices. Les fluides en évolution ont émané du magma profond en cours de cristallisation progressive, ce qui soutient la théorie selon laquelle l'or est passé du magma au réseau hydrothermal.

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[1] Contribution to the Targeted Geoscience Initiative

INTRODUCTION

The nature of mineralizing fluids is not only one of the most interesting topics in economic geology, but also an essential issue in the study of mineral deposits. Three approaches are commonly used to constrain the characteristics of mineralizing fluids: stable isotopes, fluid inclusions, and mineral chemistry (halogen contents in biotite, amphibole, apatite, etc.). Stable isotope and fluid inclusion methods are used mostly to determine the fluid source, physiochemical conditions, and evolution of mineralizing hydrothermal solutions. However, greater attention is now given to mineral chemical equilibria, which are utilized to evaluate the fluid evolution associated with magmatic rocks (Munoz and Swenson, 1981; Munoz, 1984; van Middlelaar and Keith, 1990; Lentz, 1992, 1994; Coulson et al., 2001). The principle of this method is to use internally consistent thermodynamic data for those minerals containing hydroxyl and halogens (Zhu and Sverjensky, 1991, 1992) to predict fluoride and chloride concentrations of aqueous fluids from the measured F and Cl contents in the minerals. Coulson et al. (2001) used this method in their investigation of the fluid evolution of a Au-related granitic intrusion in the Tombstone Plutonic Suite, Yukon Territory. However, this method is applicable only to equilibrium mineral assemblages. Ternary feldspar thermometry (Fuhrman and Lindsley, 1988) can test for equilibrium by assessing the extent of subsolidus recrystallization and/or exchange processes. Parts of the Lake George granodiorite stock are relatively fresh and were texturally quenched during emplacement (Seal et al., 1987; Caron, 1996; Yang et al., 2002); these equilibria may reflect the magmatic-hydrothermal conditions.

The late-stage epithermal Lake George Sb vein deposit, formerly the largest Sb producer in North America, is spatially and temporally associated with disseminated to vein-style, contact-related Au-W-Mo mineralization (Fig. 1; Scratch et al., 1984; Seal et al., 1987, 1988; Procyshyn and Morrissy, 1990; Caron, 1996). Gold anomalies are widespread in drill cores around the property (Morrissy, 1991a); ore-grade gold zones (up to 0.416 oz/t Au) in the Sb vein are locally developed (Morrissy, 1991b). Spatial and age relationships between Au mineralization and the Lake George granodiorite stock, i.e. Au-bearing quartz-carbonate vein mineralization within the stock and in the proximal metamorphic aureoles, strongly suggest that Au mineralization is genetically related to this granodiorite (Seal et al., 1988). Elsewhere in the world, gold mineralization in granitic domains has been documented to be mainly related to metaluminous, subalkalic intrusions of intermediate to felsic composition, which span the boundary between reduced ilmenite and oxidized magnetite series, in continental tectonic settings well inboard of convergent plate boundaries (Lang et al., 2000; Lang and Baker, 2001). A preliminary study of the petrochemical attributes of the Lake George granodiorite (Yang et al., 2002) indicates that they are similar to those of gold-associated granitoid rocks (Thompson et al., 1999; Lang et al., 2000; Lang and Baker, 2001). This Early Devonian granodiorite may have formed by partial melting of lower crustal amphibolitic rocks in a volcanic arc to late orogenic environment (Yang et al., 2002).

This study elaborates on the detailed mineralogy and petrology of the Lake George granodiorite. The mineral and chemical information provides geothermobarometric constraints on emplacement conditions, physiochemical conditions of crystallization, and subsequent hydrothermal fluid evolution, as well as their implications on Au mineralization.

GENERAL GEOLOGY

The Lake George granodiorite stock discordantly intruded into Silurian Kingsclear Group metasedimentary rocks, causing biotite- to cordierite-grade contact metamorphism (Seal

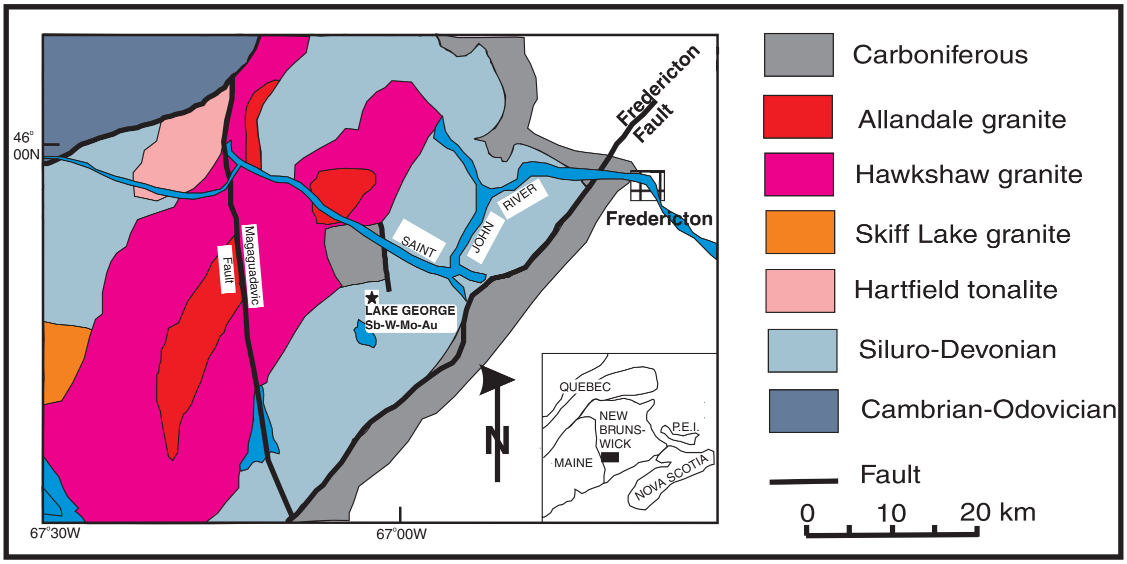

Figure 1. *Geological map of the Lake George and Pokiok area* (modified from *Procyshyn and Morrissy, 1990; McLeod et al., 1994).*

et al., 1987, 1988; Caron, 1996). Biotite occurs within the entire contact aureole whereas cordierite is restricted to the inner contact halos (Fig. 2).

The fine- to medium-grained porphyritic granodiorite consists of quartz, plagioclase, orthoclase, biotite, trace hornblende, and accessory minerals such as titanite, zircon, apatite, ilmenite, and magnetite (Fig. 3a to f). Phenocrysts 2 to 4 mm in size include subhedral plagioclase with normal zonation (Fig. 3e), euhedral to subhedral hornblende (Fig. 3a, c), and subhedral to anhedral biotite (Fig. 3b, d). Partially resorbed quartz was found in some phenocrysts. Grains in the groundmass are usually less than 1 mm in size and are composed of quartz, K-feldspar, plagioclase, biotite, and amphibole. Acicular apatite is commonly included in plagioclase, biotite, and amphibole phenocrysts. Titanite may occur as a primary phase, but mostly appears to be a secondary product of amphibole and biotite alteration, where it is associated with chlorite, magnetite, and calcite. Sericite alteration is ubiquitous in plagioclase core or along its cleavage and margin. Chlorite occurs as a common alteration phase of biotite and amphibole. Under electron probe microanalyser (EPMA) scanning mode, a number of muscovite lamellae were observed along cleavages within biotite. This muscovite is assumed to be an alteration product of the biotite. Textural relationships show that apatite and ilmenite were the earliest phases to crystallize from the melt, followed by early hornblende phenocrysts, and then by the simultaneous crystallization of biotite and plagioclase, and later, by quartz and orthoclase.

The Hawkshaw granite, a major phase of the adjacent polyphase Pokiok batholith, outcrops approximately 3 km northwest of the Lake George ore deposit (Fig. 1). It comprises mainly medium- to coarse-grained, pink K-feldspar-porphyritic biotite granite to granodiorite with minor equigranular pink biotite granite (Whalen, 1993). The Lake George granodiorite exhibits petrochemical features typical of an I-type granite, derived from the partial melting of arc-like lower continental crust in a volcanic arc to late orogenic environment, at approximately the same time as the main batholith (Yang et al., 2002). It most closely resembles the Hawkshaw phase of the batholith. However, mineralogical differences suggest a petrogenetic association linked by fractional crystallization (Yang et al., 2002).

SAMPLING METHODOLOGY AND ANALYTICAL PROCEDURES

The Lake George granodiorite stock is located 500 m north of the Hibbard shaft and lacks surface exposure, except for quartz-feldspar porphyry dykes and lamprophyre dykes (Procyshyn and Morrissy, 1990). The intrusion occurs at 350 m depth as indicated by diamond drilling (Fig. 2), although the depth to which these drillholes intersect the intrusion is shallow. Typically, the porphyritic granodiorite is weakly to moderately altered in the near-surface drill intersections, consistent with their position in the stock (see Caron, 1996). Obtaining least-altered samples was problematic, and the samples for this study were selected from the

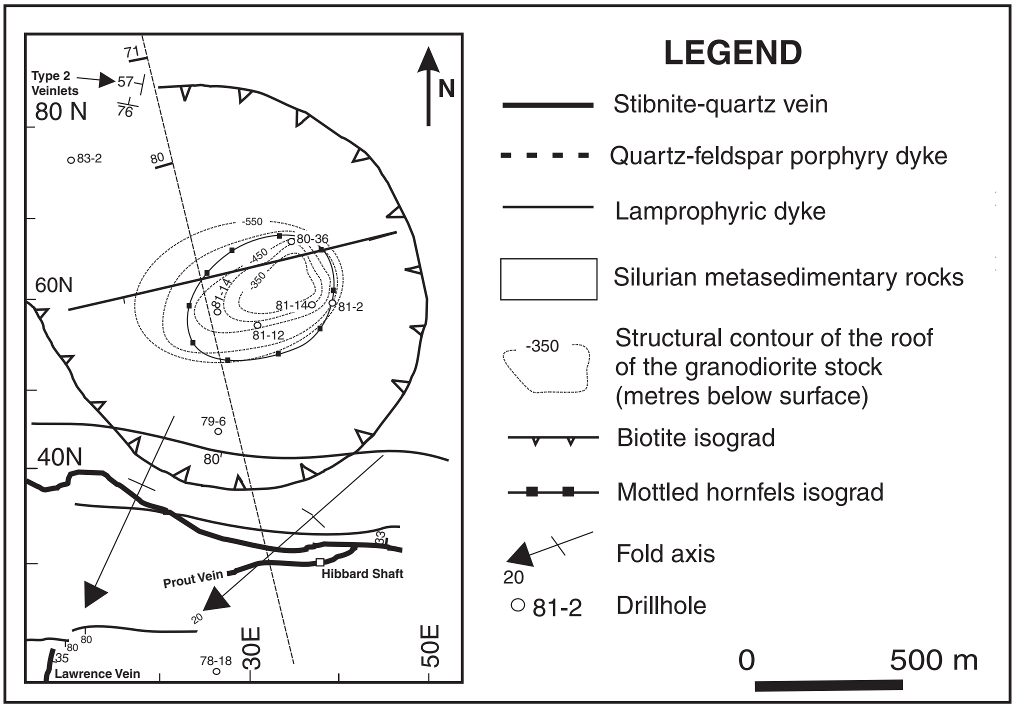

Figure 2. *Schematic geological map of the Lake George mine site (*modified from *Seal et al., 1987; Procyshyn and Morrissy, 1990). The location of the Lake George granodiorite stock is depicted as structural contours of its upper contact (in metres below the surface). Note the concentric disposition of the biotite and mottled hornfels isograds about the stock.*

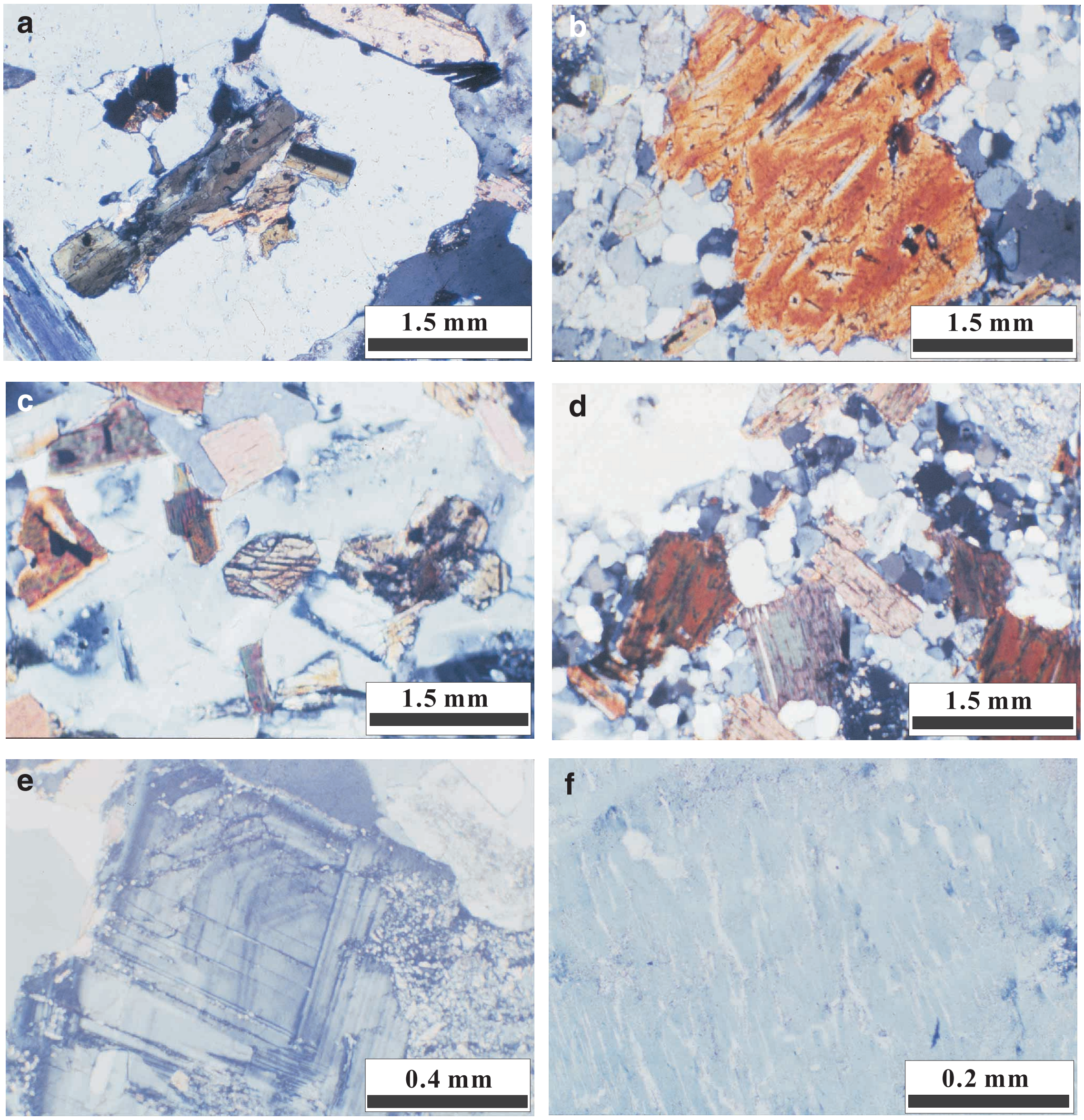

Figure 3. *Photomicrographs of the mineralogy and texture of the Lake George granodiorite.* ***a)*** *Euhedral hornblende and biotite enclosed by orthoclase; at upper right corner, euhedral titanite partly enclosed in plagioclase; crosspolarized light; sample LG83-2-2461.* ***b)*** *Subhedral biotite phenocryst in fine-grained felsic groundmass consisting of quartz, orthoclase, plagioclase, and opaques; crosspolarized light; sample LG81-12-1567.* ***c)*** *Seriate to porphyritic texture, euhedral hornblende enclosed by plagioclase; crosspolarized light; sample LG81-19-1351.* ***d)*** *Seriate to porphyritic texture, subhedral to anhedral biotite, and anhedral ilmenite (opaques); crosspolarized light; LG81-14-1886.* ***e)*** *Plagioclase phenocryst with normal zoning and polysynthetic twinning; at the lower right corner, another plagioclase is partly replaced by sericite; crosspolarized light; sample LG83-2-1995.* ***f)*** *Orthoclase with irregularly fine albite lamellae; crosspolarized light; sample LG83-2-1995.*

central parts of dykes and the deepest parts of the stock intersected by drilling, which are relatively unaltered. Ten representative granodiorite samples (50–60 cm) were collected from diamond-drill holes at the Lake George mine site for double polished thin section preparation.

Electron probe microanalyses were conducted in wavelength-dispersion mode on a JEOL-733 Superprobe at the University of New Brunswick, with 15 kV accelerating voltage, 10 nA beam current, and a maximum 40 s counting interval. The beam size was set to be 5 µm. A combination of various mineral and metal standards were used with ZAF matrix corrections by means of CITZAF version 3.03 (J.T. Armstrong, *CITZAF Version 3.03*, available at http://www2.arnes.si/~sgszmera1/others/others/mlist.html, accessed July 2002). The analytical limits are on the order of 0.05 weight per cent. In each sample, several grains of each mineral were analyzed for textural relationship and grain positions. An average of the analytical results was taken to represent the typical composition of that mineral in each sample, together with standard deviation. Formula calculations of amphibole are based on 23 oxygens, and ferric/ferrous ratios were calculated using 13-cation normalization and charge balance. Formula calculations of biotite are based on 22 oxygens, H_2O contents were calculated by stoichiometry, and ferric/ferrous ratios were computed by charge balance. The results are presented in Tables 1, 2, 3, and 4.

RESULTS AND DISCUSSION

Feldspars

Plagioclase phenocrysts occur as euhedral to subhedral grains, with normal zonation and polysynthetic twins (Fig. 3e), typical of most porphyritic rocks of granodioritic compositions. Plagioclase phenocrysts are oligoclase to andesine on average, although considerable variation exists from core to rim (Table 1; An_{58} to An_{20}). Groundmass plagioclase is typically richer in the Ab component than phenocrysts, and ranges from oligoclase to albite from core to rim (Table 1; An_{35} to An_4). Zoning of individual groundmass plagioclase grains is poorly developed, although the An component has a fairly large range (Fig. 4).

Orthoclase phenocrysts and groundmass grains have similar compositions and are relatively homogeneous (Table 2; $Or_{93.1}Ab_{6.7}An_{0.2}$), although a few microperthite grains are preserved (Fig. 3f). Two-feldspar geothermometry (Stormer, 1975) yields temperatures lower than 500°C (Fig. 4a) for coexisting orthoclase and plagioclase, reflecting extensive subsolidus recrystallization.

A practical spreadsheet program for ternary feldspar thermometry was compiled in this study, based on the formula of Fuhrman and Lindsley (1988), and was used to test for equilibrium between plagioclase and alkali feldspar. The results indicate that T_{Ab} values are always lower than 500°C, consistent with the results using the Stormer (1975) equilibria (Fig. 4a), although T_{Or} (350–668°C) and T_{An} (342–1950°C) values are more varied. These calculations, together with the plot of An-Ab-Or (Fig. 4b), clearly demonstrate that equilibrium between plagioclase and alkali feldspar has not been achieved. We interpret this result as a subsolidus or late-stage hydrothermal process, which affected the feldspars and erased the record of original equilibration.

Biotite

Biotite has an average Fe/(Fe+Mg) ratio of 0.54 ± 0.10 (Table 3; Fig. 5a) and relatively consistent high Ti and Al contents (Table 3; Fig. 5b, c). Octahedral Al against Ti in biotite (Fig. 5b) lie on the substitutional line of $R^{2+} + Ti = 2Al^{VI}$ (r = -0.57), indicating that this relationship dominates within biotite (Lentz et al., 1997). However, the substitution of Ti^{4+} in

Table 1. Electron microprobe analyses of plagioclase

Samples	LG78-18-1190	LG78-18-1190	LG80-36-1647	LG80-36-1647	LG81-12-1567	LG81-12-1567	LG81-2-1741	LG81-2-1741	LG81-19-1351	LG83-2-2461	LG83-2-2461	LG83-2-1995	LG81-14-1291	LG81-14-1886
Texture	P (n=4)	G (n=3)	P (n=6)	G (n=2)	P (n=9)	G (n=2)	P (n=3)	G (n=3)	G (n=8)	P (n=6)	G (n=1)	P (n=6)	P (n=4)	G (n=3)
SiO_2	56.9	67.3	56.7	63.7	59.7	67.0	61.5	62.7	60.9	61.0	64.5	59.2	61.1	58.9
TiO_2	0.02	0.01	0.04	0.00	0.01	0.04	0.01	0.00	0.04	0.01	0.00	0.02	0.02	0.01
Al_2O_3	26.67	19.64	26.08	21.98	24.75	20.07	23.66	22.77	23.97	24.27	22.95	25.61	24.28	24.70
Fe_2O_3	0.23	0.21	0.51	0.07	0.15	0.02	0.09	0.15	0.23	0.20	0.18	0.16	0.17	0.10
Cr_2O_3	0.01	0.00	0.01	0.01	0.00	0.00	0.01	0.00	0.00	0.01	0.00	0.00	0.01	0.01
MnO	0.02	0.02	0.00	0.01	0.01	0.02	0.01	0.01	0.01	0.01	0.02	0.01	0.01	0.00
MgO	0.04	0.01	0.22	0.06	0.04	0.00	0.02	0.01	0.01	0.00	0.00	0.00	0.00	0.00
CaO	8.10	2.48	7.44	1.43	5.17	0.48	5.36	4.15	5.54	5.89	3.81	5.65	5.82	6.67
BaO	0.04	0.01	0.07	0.05	0.03	0.04	0.01	0.03	0.06	0.04	0.00	0.02	0.06	0.09
Na_2O	6.20	9.23	6.50	9.62	8.03	11.23	8.07	9.13	7.84	7.96	9.52	7.74	8.01	7.60
K_2O	0.67	0.14	0.76	1.19	0.50	0.28	0.27	0.25	0.64	0.48	0.18	0.35	0.37	0.26
TOTAL	98.9	99.1	98.3	98.1	98.4	99.2	99.0	99.2	99.2	99.8	101.0	98.8	99.8	98.4
X_{Ab}	0.557	0.863	0.579	0.860	0.715	0.962	0.719	0.787	0.692	0.691	0.811	0.698	0.698	0.663
X_{Or}	0.040	0.009	0.047	0.070	0.029	0.016	0.016	0.014	0.038	0.027	0.010	0.021	0.021	0.015
X_{An}	0.402	0.128	0.374	0.070	0.255	0.023	0.265	0.199	0.271	0.282	0.179	0.281	0.280	0.322

Note: P = phenocryst; G = groundmass; n = number of spot analyses

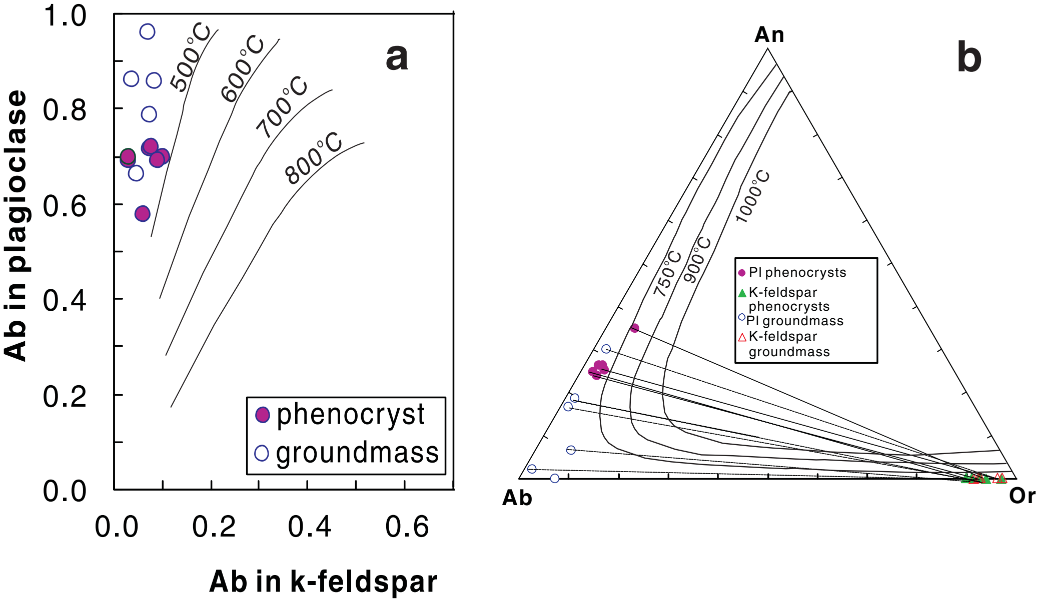

Figure 4. *Compositions of feldspars from the Lake George granodiorite.* ***a)*** *Albite contents (mole) of plagioclase versus coexisting K-feldspar. Isotherms for coexisting sanidine and plagioclase at 1 kbar are from Stormer (1975). Albite contents in plagioclase phenocrysts are lower than those in groundmass grains that have a wide variation, whereas albite contents in K-feldspar have a relatively narrow range.* ***b)*** *An-Ab-Or component plot for coexisting plagioclase and K-feldspar. The isotherms of the ternary-feldspar miscibility gap at 1 kbar are from Fuhrman and Lindsley (1988). The Ab and An components of plagioclase in groundmass have a wider variation than those in phenocrysts, where K-feldspar in both phenocrysts and groundmass grains has a relatively narrow composition range.*

Table 2. Electron microprobe analyses of K-feldspar

Samples	LG78-18-1190	LG80-36-1647	LG80-36-1647	LG81-12-1567	LG81-12-1567	LG81-2-1741	LG81-2-1741	LG79-6-612	LG81-14-1291	LG81-19-1351	LG83-2-2461	LG83-2-1995	LG81-14-1886
Texture	G (n=3)	P (n=2)	G (n=1)	P (n=4)	G (n=2)	P (n=6)	G (n=4)	G (n=4)	G (n=6)	P (n=3)	P (n=5)	P (n=6)	G (n=5)
SiO_2	64.8	64.8	64.8	63.8	65.2	64.9	65.2	64.6	65.2	64.7	64.2	64.4	65.0
TiO_2	0.05	0.03	0.12	0.03	0.03	0.02	0.02	0.04	0.18	0.02	0.01	0.01	0.02
Al_2O_3	18.36	18.81	18.47	18.51	18.35	18.32	18.24	18.16	18.38	18.65	18.41	18.46	18.79
Fe_2O_3	0.09	0.03	0.00	0.07	0.01	0.04	0.03	0.06	0.02	0.03	0.04	0.04	0.07
Cr_2O_3	0.00	0.00	0.01	0.01	0.01	0.01	0.00	0.00	0.00	0.00	0.01	0.02	0.00
MnO	0.00	0.02	0.03	0.02	0.02	0.02	0.02	0.01	0.02	0.01	0.00	0.02	0.01
MgO	0.00	0.00	0.00	0.01	0.00	0.00	0.01	0.04	0.00	0.00	0.00	0.01	0.00
CaO	0.04	0.01	0.01	0.06	0.06	0.06	0.04	0.03	0.03	0.02	0.04	0.04	0.00
BaO	0.07	0.66	0.11	0.42	0.27	0.18	0.12	0.12	0.17	0.13	0.22	0.06	0.31
Na_2O	0.39	0.65	0.93	0.78	0.76	0.84	0.81	0.80	0.31	0.34	0.98	1.12	0.51
K_2O	15.61	15.11	15.13	14.87	15.04	14.89	15.01	14.93	16.07	16.18	15.20	14.97	15.82
TOTAL	99.5	100.1	99.6	98.6	99.7	99.3	99.5	98.7	100.4	100.1	99.1	99.1	100.5
X_{Ab}	0.037	0.060	0.085	0.073	0.071	0.079	0.075	0.075	0.029	0.030	0.089	0.100	0.047
X_{Or}	0.961	0.939	0.914	0.924	0.926	0.918	0.923	0.924	0.970	0.969	0.908	0.897	0.953
X_{An}	0.002	0.001	0.001	0.003	0.003	0.003	0.002	0.001	0.002	0.001	0.002	0.002	0.000

Note: P = phenocryst; G = groundmass; n = number of spot analyses

biotite for (Fe, Mg) $^{2+}$ cannot be compensated in the way of $Ti^{VI} + 2Al^{IV} = R^{2+} + 2Si^{IV}$ (Wones, 1980) because of poor correlation (Fig. 5c).

The large range of calculated halogen fugacity ratios (f_{HF}/f_{HCl} = -2 to 0, at 400°C) for magmatic fluids in equilibrium with biotite, reflecting the significant range of log(X_F/X_{Cl}) ratios in biotite (Fig. 6), implies that the halogens in biotite were re-equilibrated with various Cl-rich magmatic fluids. These fluids probably emanated from the crystallizing granodiorite magma at depth and differentially partitioned the volatiles and solutes from that melt during progressive crystallization. The data illustrated in Figure 6 may suggest that either biotite compositions represent a crystallization sequence, an unreasonable conclusion for the Lake George granodiorite, or that biotite has been re-equilibrated with orthomagmatic to exogenetic fluids of varied composition (i.e. those with different f_{HF}/f_{HCl} ratios). If magmatic, these fluids could have changed with time as the magma cooled and solidified at depth, so that the f_{HF}/f_{HCl} ratios of the fluids would increase progressively.

Low calculated $Fe^{3+}/(Fe^{2+}+Fe^{3+})$ ratios in both biotite (average 0.06; Table 3), which falls into the field between NNO and QFM buffer in the Fe^{3+}-Fe^{2+}-Mg ternary diagram of Wones and Eugster (1965) (not shown), and amphibole (average 0.07; Table 4), the presence of ilmenite as the predominant Fe-Ti oxide, and low magnetic susceptibility (average 6 × 10^{-5} SI), indicate relatively reducing conditions for the magma and associated magmatic fluids. Reducing conditions favour the transfer of Au from the melt to fluid systems if sulphur concentration is low in the magma (Thompson et al., 1999; Lang et al., 2000; Lang and Baker, 2001), and can also play an important role in the formation of large W deposits (*see* van Middelaar and Keith, 1990).

Table 3. Electron microprobe analyses of biotite

Samples	LG78-18-1190	LG78-18-1190	LG79-6-612	LG80-36-1647	LG80-36-1647	LG81-2-1741	LG81-2-1741	LG81-12-1567	LG81-14-1291	LG81-14-1886	LG81-19-1351	LG83-2-1995	LG83-2-1995	LG83-2-2461
Texture	P (n=3)	G (n=1)	P (n=3)	P (n=2)	G (n=3)	P (n=5)	G (n=3)	P (n=7)	P (n=4)	P (n=6)	G (n=4)	P (n=3)	G (n=4)	P (n=3)
SiO_2	36.5	37.4	35.6	34.7	35.3	35.9	39.5	35.9	36.2	36.8	36.3	36.0	36.2	36.1
TiO_2	4.00	4.14	4.07	3.82	3.40	4.21	2.59	4.00	4.01	3.91	4.23	3.91	4.01	3.98
Al_2O_3	15.43	15.17	15.51	14.97	16.07	14.82	15.72	15.69	15.89	15.44	15.23	15.14	14.95	15.18
Cr_2O_3	0.02	0.01	0.03	0.02	0.01	0.00	0.03	0.02	0.02	0.02	0.00	0.01	0.03	0.01
Fe_2O_3	1.91	3.53	3.62	3.91	1.88	1.86	0.33	2.96	7.56	3.55	2.50	1.67	0.70	0.27
FeO	15.45	13.82	18.28	16.18	18.21	19.65	21.38	18.92	11.57	17.47	20.18	20.58	21.41	21.86
MnO	0.40	0.47	0.25	0.40	0.29	0.37	0.31	0.38	0.21	0.39	0.35	0.39	0.44	0.38
MgO	12.26	12.33	9.74	9.40	9.51	9.47	9.74	9.59	11.53	10.23	9.01	9.69	9.41	8.71
CaO	0.02	0.04	0.01	0.11	0.59	0.04	0.13	0.06	0.06	0.04	0.18	0.10	0.03	0.49
BaO	0.26	0.02	0.10	0.18	0.16	0.26	0.00	0.11	0.22	0.12	0.13	0.11	0.16	0.10
Na_2O	0.11	0.14	0.08	0.08	0.04	0.12	0.08	0.11	0.08	0.10	0.11	0.08	0.08	0.09
K_2O	8.93	9.04	8.07	8.70	8.32	8.84	5.85	8.07	8.68	8.99	8.89	8.50	9.15	8.90
H_2O	3.88	3.86	3.75	3.62	3.78	3.82	4.01	3.80	3.61	3.83	3.84	3.85	3.89	3.89
Cl	0.06	0.05	0.06	0.04	0.05	0.08	0.05	0.06	0.05	0.03	0.04	0.02	0.04	0.04
F	0.15	0.30	0.31	0.35	0.13	0.12	0.00	0.24	0.81	0.31	0.20	0.14	0.04	0.00
-O=Cl	0.01	0.01	0.01	0.01	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
-O=F	0.06	0.13	0.13	0.15	0.06	0.05	0.00	0.10	0.34	0.13	0.08	0.06	0.02	0.00
Total	99.3	100.2	99.3	96.3	97.7	99.5	99.7	99.8	100.2	101.1	101.1	100.2	100.4	99.9
^{IV}Si	5.51	5.57	5.45	5.49	5.48	5.53	5.89	5.48	5.42	5.53	5.510	5.505	5.532	5.544
^{IV}Al	2.489	2.429	2.552	2.511	2.516	2.474	2.112	2.523	2.580	2.469	2.490	2.495	2.468	2.456
T site	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000
^{VI}Al	0.258	0.237	0.250	0.277	0.424	0.216	0.647	0.294	0.223	0.266	0.233	0.231	0.229	0.293
Ti	0.455	0.464	0.470	0.454	0.397	0.487	0.290	0.459	0.451	0.442	0.483	0.449	0.462	0.460
Cr	0.003	0.001	0.003	0.002	0.001	0.000	0.004	0.002	0.002	0.002	0.000	0.001	0.003	0.002
Fe^{3+}	0.085	0.154	0.165	0.183	0.079	0.078	0.013	0.131	0.393	0.156	0.106	0.074	0.028	0.010
Fe^{2+}	2.109	2.009	2.642	2.470	2.530	2.690	2.691	2.660	2.001	2.485	2.773	2.768	2.800	2.834
Mn	0.052	0.059	0.032	0.053	0.039	0.049	0.039	0.049	0.027	0.050	0.045	0.050	0.056	0.050
Mg	2.760	2.741	2.226	2.214	2.203	2.174	2.163	2.179	2.572	2.291	2.039	2.207	2.145	1.994
O site	5.722	5.666	5.789	5.653	5.671	5.694	5.846	5.773	5.669	5.691	5.679	5.781	5.724	5.643
Ca	0.003	0.006	0.002	0.019	0.099	0.007	0.021	0.009	0.009	0.006	0.028	0.017	0.005	0.081
Ba	0.015	0.001	0.006	0.011	0.010	0.016	0.000	0.007	0.013	0.007	0.008	0.007	0.009	0.006
Na	0.033	0.040	0.025	0.023	0.013	0.035	0.023	0.031	0.022	0.029	0.032	0.025	0.022	0.025
K	1.720	1.720	1.576	1.752	1.646	1.736	1.111	1.570	1.656	1.722	1.722	1.655	1.785	1.743
A site	1.772	1.768	1.609	1.805	1.768	1.794	1.155	1.618	1.699	1.765	1.790	1.703	1.822	1.856
OH^-	3.915	3.846	3.835	3.817	3.921	3.922	3.987	3.869	3.607	3.844	3.894	3.926	3.972	3.990
Cl	0.015	0.013	0.016	0.011	0.014	0.020	0.013	0.015	0.012	0.008	0.011	0.006	0.009	0.009
F	0.070	0.141	0.150	0.173	0.065	0.058	0.000	0.116	0.381	0.148	0.095	0.068	0.019	0.001
Mg/(Mg+Fe)	0.558	0.559	0.442	0.455	0.458	0.440	0.444	0.439	0.518	0.465	0.415	0.437	0.431	0.412
$Fe^{3+}/(Fe^{3+}+Fe^{2+})$	0.039	0.071	0.059	0.069	0.030	0.028	0.005	0.047	0.164	0.059	0.037	0.026	0.010	0.004

Note: P = phenocryst; G = groundmass; n = number of spot analyses

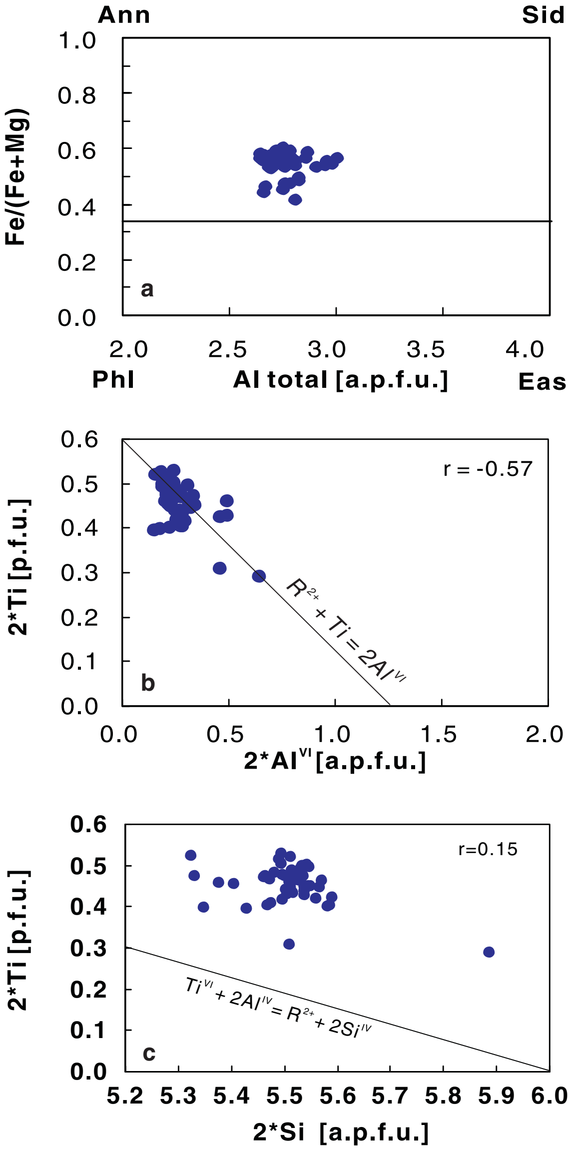

Figure 5. *Compositions of biotite from the Lake George granodiorite.* ***a)*** *Fe/(Fe+Mg) versus Al(total) biotite compositions on annite-eastonite-phlogopite-siderophyllite diagram.* ***b)*** *Plot of Al^{VI} versus Ti in biotite illustrating its fit along the substitutional line of $R^{2+} + Ti = 2Al^{VI}$ (r = -0.57).* ***c)*** *Ti^{VI} versus Si^{IV} contents of biotite. The poor correlation (r = 0.15) indicates that the substitution of Ti in biotite (Fe^{2+}, Mg) was not compensated by $Ti^{VI} + 2Al^{IV} = R^{2+} + 2Si^{IV}$. p.f.u. = per formula unit; a.p.f.u. = atoms per formula unit*

Amphibole

According to the classification of Leake et al. (1997), the amphibole in the Lake George granodiorite is a calcic hornblende. Mineral formulae are based on 23 oxygens and standardized on 13 cations (excluding Ca, Na, and K) (Table 4). The calcic hornblende in the granodiorite has Ca in the site of M4 >1.0 with Na<0.5, Si/(Si+Al+Ti)>0.775, and average Fe/(Fe+Mg) of 0.53 ± 0.06 (Table 4; Fig. 7), and moderate Al_2O_3 (<10 wt. %) and TiO_2 (<2 wt. %) contents (Table 4). It is notable that Fe/(Fe+Mg) distribution coefficients $[Fe]K_D^{Hb/Bi}$ (Lentz, 1994) between coexisting amphibole and biotite are about 0.20 to 0.68, suggesting that Fe preferentially partitions into biotite during magma crystallization.

Al-in-amphibole geobarometry (Hammarstrom and Zen, 1986) has been widely used to estimate the pressures of igneous amphibole crystallization with assemblages of quartz, plagioclase, orthoclase, biotite, amphibole, titanite, and ilmenite or magnetite. The method has been tested with natural mineral and experimental data for pressures ranging from < 1 to 10 kbar (Rutter et al., 1989; *see* Stein and Dietl, 2001, and references therein). Al-in-amphibole geobarometry (Hammarstrom and Zen, 1986) yields crystallization pressures for the Lake George granodiorite ranging from 3 to 5 kbar, significantly higher than that of the final emplacement depth of the stock (<2 kbar), based qualitatively on texture and Ab-Or-Q-H_2O phase equilibria (Yang et al., 2002). This pressure discrepancy is interpreted to be the result of early amphibole crystallization prior to the final emplacement of the stock, implying that the magma became water-saturated during ascent.

Blundy and Holland (1990) established an empirical amphibole-plagioclase geothermometer based on the edenite-tremolite reaction (e.g. edenite + 4 quartz = tremolite + albite), which could be used to estimate the temperature of quartz-bearing intermediate to felsic igneous rocks with plagioclase (An≤0.92) and Si in amphibole (≤7.8 atoms per formula unit (a.p.f.u.)). The geothermometer is described as the following formula.

$$T[\pm 311K] = \frac{0.677P[kb] - 48.98}{-0.0429 - 0.0083144\ln\left\{\left(\frac{Si-4}{8-Si}\right)X\frac{Pl}{Ab}\right\}}$$

where Si represents atoms per formula unit in amphibole and $X\frac{Pl}{Ab}$ denotes the mole fraction of albite in plagioclase. The samples from the Lake George granodiorite (Tables 1 and 4)

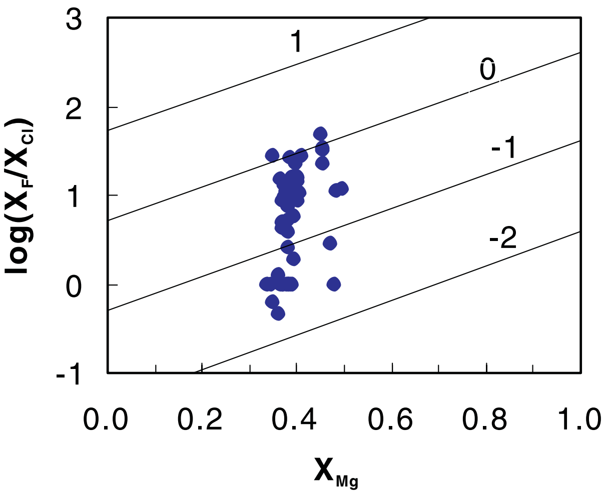

Figure 6.

Diagram X_{Mg} versus $\log(X_F/X_{Cl})$ of biotite from the Lake George granodiorite; X_F and X_{Cl} are the mole fractions of F and Cl in the hydroxyl site, respectively. Contours are the logarithm of the fluorine-chlorine fugacity ratios log (f_{HF}/f_{HCl}) for a fluid in equilibrium with biotite (Zhu and Sverjensky, 1992), calculated at 400°C.

Table 4. Electron microprobe analyses of amphibole

Samples	LG80-36-1647		LG81-19-1351		LG81-2-1741		LG83-2-1995		LG83-2-2461	
	1 grain (n=2)		5 grains (n=19)		1 grain (n=3)		1 grain (n=5)		1 grain (n=3)	
	x	1 σ	x	1 σ	x	1 σ	x	1 σ	x	1 σ
SiO_2	44.2	0.7	44.4	1.7	45.0	0.9	44.7	1.8	44.3	0.3
TiO_2	1.52	0.14	1.53	0.42	1.33	0.20	1.27	0.37	1.58	0.10
Al_2O_3	9.61	0.45	9.61	1.16	9.23	0.50	8.20	1.58	9.48	0.28
Cr_2O_3	0.02	0.01	0.04	0.05	0.03	0.02	0.02	0.01	0.01	0.02
Fe_2O_3	2.66	0.01	2.70	0.04	2.67	0.02	2.68	0.04	2.71	0.03
FeO	17.34	0.65	16.88	1.03	15.20	1.42	17.44	0.62	16.99	0.38
MgO	8.96	0.23	9.43	1.03	10.93	1.28	10.18	0.82	9.44	0.08
CaO	11.21	0.03	10.84	0.94	11.51	0.11	11.29	0.23	11.22	0.10
BaO	0.04	0.06	0.02	0.05	0.02	0.03	0.04	0.05	0.02	0.03
Na_2O	1.44	0.01	1.38	0.36	1.64	0.02	1.49	0.22	1.58	0.03
K_2O	0.98	0.08	0.74	0.21	0.72	0.28	0.76	0.21	0.96	0.05
Cl	0.04	0.01	0.03	0.01	0.03	0.01	0.02	0.01	0.03	0.01
F	0.00	0.00	0.03	0.05	0.09	0.11	0.03	0.07	0.00	0.00
-O=F,Cl	0.01	0.00	0.02	0.02	0.05	0.05	0.02	0.03	0.01	0.00
Total	98.8		98.4		98.9		99.0		99.1	
Si	6.64	0.10	6.67	0.22	6.68	0.08	6.71	0.26	6.63	0.05
Al^{IV}	1.362	0.097	1.331	0.219	1.322	0.082	1.289	0.256	1.367	0.049
Sum T	8.000		8.000		8.000		8.000		8.000	
Al^{VI}	0.341	0.014	0.369	0.106	0.293	0.025	0.164	0.048	0.305	0.004
Ti	0.172	0.016	0.173	0.047	0.149	0.023	0.144	0.042	0.178	0.010
Fe^{3+}	0.163	0.001	0.165	0.002	0.162	0.002	0.163	0.003	0.165	0.001
Cr	0.002	0.000	0.004	0.006	0.004	0.001	0.002	0.001	0.002	0.003
Mg	2.005	0.050	2.112	0.238	2.415	0.267	2.278	0.173	2.106	0.026
Fe^{2+}	2.205	0.057	2.085	0.155	1.905	0.221	2.134	0.098	2.149	0.035
Mn	0.110	0.014	0.090	0.009	0.072	0.013	0.111	0.010	0.094	0.013
Ba	0.002	0.003	0.001	0.003	0.001	0.002	0.003	0.003	0.001	0.002
Sum C	5.000		5.000		5.000		5.000		5.000	
Fe^{2+}	0.147	0.029	0.213	0.196	0.150	0.027	0.224	0.021	0.151	0.011
Ca	1.806	0.001	1.730	0.172	1.829	0.020	1.775	0.021	1.799	0.013
Na	0.048	0.028	0.057	0.052	0.021	0.020	0.001	0.003	0.050	0.024
Sum B	2.000		2.000		2.000		2.000		2.000	
Na	0.386	0.052	0.401	0.104	0.470	0.003	0.434	0.067	0.457	0.009
K	0.189	0.017	0.142	0.039	0.138	0.054	0.145	0.040	0.183	0.007
Sum A	0.575		0.543		0.608		0.579		0.639	
$Fe^{3+}/(Fe^{3+}+Fe^{2+})$	0.065	0.008	0.067	0.005	0.073	0.007	0.065	0.023	0.067	0.024

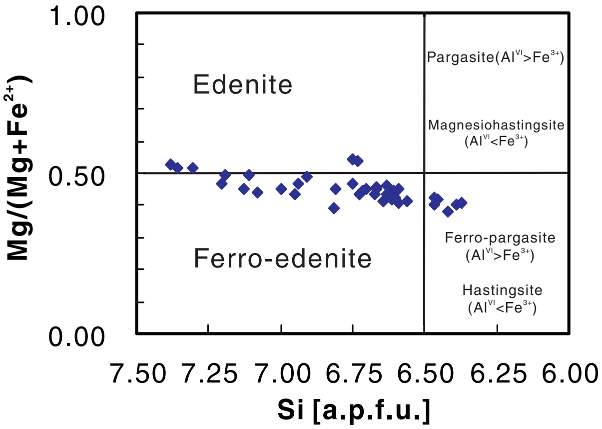

Figure 7.

Compositions of calcic amphibole from the Lake George granodiorite plotted in the amphibole classification diagram in terms of Si [a.p.f.u.] and Mg/(Mg + Fe^{2+}) by Leake et al. (1997). a.p.f.u. = atoms per formula unit

meet the requirements of the geothermometer. According to the above equation, temperatures calculated for cogenetic amphibole and plagioclase in five granodiorite samples indicate crystallization between 729°C and 772°C, which is near the wet granodiorite solidus at 3 to 5 kbar (Whitney, 1988).

Although the amphibole-plagioclase geothermometer (Blundy and Holland, 1990) is controversial (Hammarstrom and Zen, 1992; Poli and Schmidt, 1992; Rutherford and Johnson, 1992), it can produce reasonable temperatures if the requirements of the geothermometer are met (*see* Stein and Dietl, 2001). Its application to the Lake George granodiorite also yields reasonable temperature estimates. However, Holland and Blundy (1994) revised their geothermometer, taking into consideration all the components involved in the edenite-tremolite reaction. The revised equations were tested by Stein and Dietl (2001) and produced temperatures 30°C to 70°C lower than those of the original calibration, but with significantly larger errors. Hence, we chose to use the original equation by Blundy and Holland (1990).

Apatite

Apatite is a common accessory mineral in the granodiorite stock and occurs as acicular crystals ranging from 10 to 200 µm in length. It is commonly enclosed by plagioclase, amphibole, and biotite, and rarely by quartz. Interestingly, fluid inclusions with vapour/fluid ratios ranging from 1/1 to 3/1 were found in the apatite, indicating that it crystallized from a vapour-saturated wet melt at an early stage.

As the Lake George granodiorite is metaluminous to weakly peraluminous (A/CNK = 0.99–1.13) and calc-alkaline (Yang et al., 2002), the temperature of apatite crystallization can be estimated using the equation given by Piccoli et al. (1999).

$$T = \frac{[26400 C^{1} SiO_2 - 4800]}{[124 \cdot C^{1} SiO_2 - \ln(C^{1} P_2 O_5) - 3.97]}$$

where T is the apatite saturation temperature (in °K), and $C^{l}SiO_2$ and $C^{l}P_2O_5$ represent the concentrations of silica and phosphorus in the melt when apatite began to crystallize. Using the whole-rock concentrations of SiO_2 and P_2O_5 (Yang et al., 2002) as estimation of the initial melt composition, calculated apatite saturation temperatures range from 881°C to 911°C, with an average of 902 ± 14°C (n = 9), which may represent the prevolatile saturation melt temperature. It is notable that sample LG81-14-1291, which is obviously altered (low SiO_2 content of 62.4 wt. %, Yang et al., 2002; Table 1), yields an unrealistic low temperature (848°C)

Titanite

Although titanite is a normal accessory mineral in the granodiorite, most of it is secondary, formed in association with chlorite and magnetite by alteration of amphibole and ilmenite. Only a few primary grains exist. However, the compositions of primary and secondary titanite are indistinguishable.

Fe-Ti oxides

Subhedral and anhedral magnetite occurs as alteration products of amphibole and biotite, with very uncommon cubic grains found as inclusions in amphibole. Generally, magnetite is less abundant than ilmenite, which occurs separately as subhedral and anhedral inclusions in biotite and amphibole. Later ilmenite also formed during replacement of biotite, where it is usually distributed along the cleavage planes of the biotite, together with secondary chlorite or muscovite.

The drill-core samples have low magnetic susceptibility (4 x 10^{-5} SI to 21 x 10^{-5} SI, average 6 x 10^{-5} SI), consistent with low magnetite content (Ishihara, 1981) and reflecting the low $f(O_2)$ character of the granodiorite intrusion.

CONCLUSIONS

1. The crystallization sequence of rock-forming minerals is hornblende, biotite and plagioclase, followed by quartz and orthoclase. Hornblende began to crystallize at 5 kbar (16.5 km) during magma ascent; however, the final emplacement of the granodiorite stock was epizonal (less than 2 kbar).

2. The geothermometry used here yields a wide range of temperatures for the Lake George granodiorite: near liquidus (881–911°C), major crystallization (729–772°C), and subsolidus processes (<500°C). Temperatures estimated from coexisting ternary feldspars indicate that the primary igneous system was partially reset by late-stage hydrothermal subsolidus processes.
3. Late-stage magmatic fluids equilibrated with biotite are characterized by Cl-rich compositions, a large variation in halogen fugacity ratios [$\log(f_{HF}/f_{HCl})$], and reducing conditions. Evolving fluids emanated from the progressively crystallizing magma at depth, favouring Au partitioning from the magma to the hydrothermal system.

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