Report

Chemical composition and K–Ar age of Phengite from Barrovian metapelites, Loch Leven, Scotland

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Abstract: New K–Ar ages of phengite concentrates from the Barrovian metapelites of the Loch Leven area, Scotland yield 398 ± 10 , 406 ± 11 , 405 ± 11 , 400 ± 11 and 445 ± 12 Ma. All the phengites have homogeneous mineral compositions, and the metamorphic temperatures are below the closure temperature of the K–Ar phengite system; this suggests that the time of metamorphic recrystallization was between the significant age-gap of ca. 445 and ca. 400 Ma. Integration of our K–Ar ages with previous geological and geochronological data from the Loch Leven area and from the Moine Thrust, which is the major lower boundary of the Barrovian metamorphic belt, suggests that the lower boundary is present as the Fort William Slide in the Loch Leven area.

Keywords: Barrovian, K–Ar age, Phengite

Introduction

The formation and exhumation of a regional metamorphic belt is one of the most important processes in the development of an orogenic belt (Dewey and Bird, 1970; Matsuda and Ueda, 1971; Maruyama, 1997; Agard et al., 2009). In order to understand exhumation tectonics, it is critical to identify the upper and lower boundaries of a metamorphic belt, because these control

Fig. 1. Simplified geological map of the Caledonian orogenic belt of Scotland, including Barrovian metamorphic zones, major terranes and tectonic boundaries, and the location of Fig. 2 (modified after Barrow, 1893; Hutchison & Oliver, 1998). NHT = Northern Highland Terrane, GT = Grampian Terrane, MT = Moine Thrust, GGF = Great Glen Fault, HBF = Highland Boundary Fault.

the geological structure and exhumation channel of the metamorphic belt (e.g. Kawai et al., 2006, 2007; Aoki et al., 2009, 2011).

The Barrovian metamorphic belt in the Caledonides of Scotland is situated between the Moine Thrust and the Highland Boundary Fault (Fig. 1). The Moine Thrust is well known as the lower boundary of the Barrovian metamorphic belt in the Northern Highland terrane (e.g. Strachan et al., 2002; Butler, 2004, 2010), but until now, however, there are no reports of the presence of the lower boundary in the Grampian terrane. Based on the geological structure of the Loch Leven area (Roberts, 1976; Roberts and Treagus, 1977), it remains possible that the lower tectonic boundary occurs there. The metamorphic rocks below the bottom boundary of a

Fig. 2. Simplified geological map of the Loch Leven area (modified after Roberts and Treagus, 1977), showing the complete Dalradian stratigraphy, the position of the two main tectonic slides, a schematic cross-section through this area, and sample localities.

regional metamorphic belt predictably have younger K–Ar ages compared with those above the thrust boundary (e.g. Aoki et al., 2008, 2011). In order to test this prediction, we have analyzed the composition of phengites in metapelites from the Loch Leven area, and have dated phengite concentrates by the un-spiked K–Ar method.

Geological outline

The Barrovian metamorphic belt is divisible into two subunits: the Moine Supergroup and the Dalradian Supergroup based on their original stratigraphic relations. The Dalradian Supergroup, which is structurally and stratigraphically above the Moine Supergroup, is mainly composed of three Groups from structural bottom to top: the Appin, Argyll, and Southern Highland (e.g. Anderson, 1985). The Moine Supergroup is composed of six Groups from structural bottom to top: the Morar, Glenfinnan, Loch Eil Groups, the East Sutherland Moine, the Glen and Dava Successions, and the Grampian Group. These groups are mainly composed of passive continental margin turbiditic meta-sediments (e.g. Strachan et al., 2002).

The Barrovian metamorphic zones are defined by the occurrence of index minerals in metapelites: chlorite, biotite, garnet, staurolite, kyanite and sillimanite, in ascending order of metamorphic grade from greenschist facies through amphibolite to granulite facies (e.g. Barrow, 1893, 1912; Winchester, 1974; Atherton, 1977; Fettes et al. 1985) (Fig. 1). The age of the Barrovian metamorphism is estimated to be ca. 480–430 Ma, based on ^{40}Ar – ^{39}Ar , K–Ar, Rb–Sr and Sm–Nd dating

(e.g. Dempster, 1985; Elias et al. 1988; Kelley, 1988; Dallmeyer et al. 2001; Oliver, 2001).

The rocks in the Loch Leven area belong to the Appin and Grampian Groups, and comprise twelve stratigraphic types from structural bottom (the Eilde Flags) to top (the Appin Phyllite) (Roberts, 1976; Roberts and Treagus, 1977) (Fig. 2); lithological details of each are provided by Treagus (1974) and Roberts and Treagus (1977). In the Loch Leven area the rocks have been deformed in the chlorite-biotite zones (ca. 4–5 kbar and ca. 300–500°C; Atherton, 1977) and have been deformed by three recumbent folds, the Appin Syncline, Kinlochleven Anticline and Ballachuish Syncline (highest), which all face towards the NW (Treagus, 1974, 1987; Roberts, 1976; Roberts and Treagus, 1977; Hambrey et al., 1991). The lower limbs of the two synclines are occupied by the Fort William and Ballachulish Slides. In this study, we analyzed phengite concentrates for their K–Ar ages from six metapelites (Ballachulish Slate: sample no. LL13, 46; Leven Schist: LL19, 20; Binnein Schist: LL16; Eilde Flags: LL24).

Sample descriptions and phengite compositions

The chemical composition of phengite was determined with a JEOL JXA-8800M electron probe micro-analyzer (EPMA) at the Tokyo Institute of Technology. Operating conditions were in WDS mode with 15 kV accelerating voltage, a beam current of 12 nA, and a probe diameter focused to < 1 µm. We used natural and synthetic silicate and oxide standards for calibration, and the ZAF method for matrix correction.

Fig. 3. Photomicrographs showing representative phengite concentrates in thin section; phengite is generally present as aggregates of fine-grained crystals.

Table 1. Representative chemical compositions of phengites within the Loch Leven metapelites.

Sample no.	LL13	LL13	LL46	LL46	LL19	LL19	LL20	LL20	LL24	LL24	LL16	LL16
Type	BaS	BaS	BaS	BaS	LS	LS	LS	LS	EF	EF	BS	BS
SiO ₂	49.19	48.85	49.88	49.95	48.22	46.97	47.61	48.15	49.73	48.53	48.05	48.09
TiO ₂	0.34	0.33	0.41	0.24	0.38	0.48	0.41	0.36	0.37	0.38	0.36	0.35
Al ₂ O ₃	31.86	33.98	31.76	31.87	31.25	31.37	32.68	31.90	32.31	32.37	30.17	30.34
Cr ₂ O ₃	0.00	0.00	0.08	0.00	0.00	0.10	0.00	0.00	0.00	0.02	0.04	0.04
FeO*	2.62	2.27	1.97	2.13	3.89	4.30	4.20	4.37	2.80	2.57	5.23	4.39
MnO	0.03	0.00	0.00	0.01	0.00	0.02	0.00	0.01	0.03	0.02	0.04	0.01
MgO	1.53	1.23	2.08	2.26	1.77	1.63	1.54	1.76	1.94	1.71	1.67	1.90
CaO	0.00	0.02	0.00	0.00	0.01	0.04	0.00	0.00	0.01	0.00	0.01	0.00
Na ₂ O	0.37	0.43	0.29	0.36	0.32	0.45	0.61	0.62	0.44	0.63	0.18	0.63
K ₂ O	9.88	10.06	9.75	9.68	9.90	9.76	9.92	9.93	10.08	9.78	10.56	9.84
Total	95.82	97.17	96.21	96.49	95.73	95.12	96.96	97.10	97.70	96.00	96.29	95.59
<i>Cations (O): 11</i>												
Si	3.252	3.183	3.269	3.265	3.219	3.170	3.149	3.182	3.232	3.208	3.226	3.230
Ti	0.017	0.016	0.020	0.012	0.019	0.024	0.020	0.018	0.018	0.019	0.018	0.018
Al	2.483	2.609	2.453	2.455	2.459	2.495	2.547	2.484	2.475	2.521	2.387	2.401
Cr	0.000	0.000	0.004	0.000	0.000	0.005	0.000	0.000	0.000	0.001	0.002	0.002
Fe ²⁺	0.145	0.124	0.108	0.116	0.217	0.243	0.232	0.242	0.152	0.142	0.294	0.247
Mn	0.001	0.000	0.000	0.001	0.000	0.001	0.000	0.000	0.002	0.001	0.002	0.000
Mg	0.151	0.120	0.203	0.220	0.176	0.164	0.152	0.173	0.188	0.168	0.167	0.191
Ca	0.000	0.001	0.000	0.000	0.000	0.003	0.000	0.000	0.001	0.000	0.001	0.000
Na	0.047	0.054	0.037	0.045	0.042	0.059	0.078	0.080	0.055	0.081	0.023	0.082
K	0.833	0.836	0.815	0.807	0.843	0.840	0.837	0.837	0.836	0.825	0.904	0.843

*Total Fe as FeO. BaS: Ballachulish Slate, LS: Leven Schist, EF: Eilde Flags, BS: Binnein Schist

The investigated samples are mainly composed of quartz, plagioclase, phengite, chlorite, and epidote. Foliations and mineral lineations are defined by preferred orientations of phengite and chlorite. Phengite occurs as aggregates of fine-grained crystals of 1–10 μm in width (Fig. 3). Table 1 lists representative chemical compositions of phengite. Figure 4 shows compositional variations of the phengites in terms of their Si–Al and Si–Mg/(Mg + Fe) space. Variation of Si contents in each sample is about 0.1. Phengites in metapelites plot near

the ideal celadonite substitution line (Fig. 4A). (Mg/(Mg + Fe)) values range from 0.37 to 0.66 (Fig. 4B), and there is no systematic difference between phengites within individual samples.

K–Ar analysis

The phengites were handpicked and/or tapped from the samples after they had been crushed, sieved (60–80 mesh size) and magnetically separated. The separates were leached with diluted HNO₃ to decompose any as-

Fig. 4. Compositional variation of phengites in the Loch Leven metapelites. (A) Si–Al and (B) Si–Mg/(Mg + Fe). Cation numbers are calculated for 11 oxygens.

sociated chlorite and organic materials. Potassium concentrations were determined on a Zeeman atomic absorption spectrometer (Hitachi Z-5010) using no buffer with an analytical error within 2% at a 2 σ confidence level (Sato et al., 2005a). Digestions and chemical handling of the samples were performed in a clean laboratory to minimize the blank level. Ar isotope ratios were measured on a sector-type noble gas mass spectrometer (GVI-5400, GV Instruments), in JAMSTEC. For age calculations, fractionation effects for initial $^{40}\text{Ar}/^{36}\text{Ar}$ ratios were uncorrected with the measured $^{38}\text{Ar}/^{36}\text{Ar}$, because expected ages are older than 1 Ma. The Ar isotope ratios and Ar contents were normalized through repeated measurements of atmospheric Ar, and HD-B1 and SORI93 biotite standards (Sato et al., 2005b). More detailed descriptions are given in Sato et al. (2005a, b).

K–Ar ages were determined as 398 ± 10 (sample no. LL46), 406 ± 11 (LL20), 405 ± 11 (LL19), 400 ± 11 (LL24), 340 ± 9 Ma (LL13) and 445 ± 12 Ma (LL16) (Table 2). These errors are reported to 1 sigma. In some low-grade metamorphic rocks (< 300°C), phengite

shows a wide compositional variation, suggesting a mixture of detrital and metamorphic components (e.g. Itaya and Fujino, 1999; Takeshita et al., 2004). Phengites in our samples are aligned in the schistosity and have a chemical composition that is particular to each sample (Figs. 3, 4). The inferred metamorphic temperatures are below the closure temperature of the K–Ar phengite system (Villa, 1998). Hence, all the analyzed phengites are interpreted to be metamorphic, and the K–Ar ages should represent the time of phengite recrystallisation.

The 445 ± 12 Ma age of the Binnein Schist (LL16) is consistent with reported muscovite K–Ar ages of the relevant Barrovian metamorphic rocks (ca. 480–430 Ma; e.g. Kelley, 1988; Oliver, 2001). This is consistent with the idea that the age documents the time of phengite formation during ‘Barrovian metamorphism’. According to Roberts and Treagus (1977) and Hambrey et al. (1991), the Glen Coe Quartzite and the Eilde Schist are lithologically very similar to the Eilde Quartzite and Binnein Schist respectively. Integration of these details with our data indicates that the lithologies from the Glen Coe Quartzite to the Eilde Quartzite are coherent and recrystallized at about the same time at ca. 445 Ma.

Our K–Ar ages of ca. 406–400 Ma obtained from the Ballachulish Slate (LL46), Leven Schist (LL19, 20) and Eilde Flags (LL24) are significantly younger than those of the Binnein Schist (445 Ma) and of the main Barrovian metamorphic rocks (ca. 480–430 Ma). Nonetheless, it is noteworthy that there is no significant difference in their K–Ar ages regardless of the variation of their K_2O contents (Table 2). Hence, it would be fair to say that the K–Ar ages of ca. 406–400 Ma are geologically meaningful, and that they show the timing of phengite formation during metamorphism. One sample (LL13) of Ballachulish Slate has an anomalously young age of 340 ± 9 Ma. Because low-K samples ($\text{K}_2\text{O} < 1.5$ wt.%) generally showed relatively younger K–Ar ages with large uncertainties compared to those of high-K samples (e.g. Itaya and Takasugi, 1988; Hirajima et al., 1992), such younger age obtained from LL13 is probably due to impurity of phengite concentrates by mixture of plagioclase.

The Eilde Flags are stratigraphically below the Binnein Schist and yet they also have younger K–Ar ages (Fig. 2). Moreover, the K–Ar age of the Eilde Flags is closer to the age of movement on the Moine Thrust (ca. 437–408 Ma; Freeman et al., 1998; Goodenough et al., 2011), and the Fort William Slide bounds the Eilde Flags and overlying Formations (Fig. 2). Considering these data and geologic relations, the Eilde Flags are

Table 2. K–Ar data for phengites separated from metapelites of the Loch Leven area.

sample	weight (g)	K ₂ O (wt%)	error	⁴⁰ Ar rad (cc/g)	error	Atmospheric contamination (%)	Age (Ma)	error (1σ)
LL46 (BaS)	0.00498	3.206	0.064	4.612E-05	7.750E-07	6.71	398.2	10.4
LL13 (BaS)	0.00410	1.304	0.026	1.575E-05	2.624E-07	5.97	340.1	8.9
LL20 (LS)	0.00424	3.196	0.064	4.703E-05	7.835E-07	5.06	406.4	10.6
LL19 (LS)	0.00613	4.146	0.083	6.082E-05	1.013E-06	3.51	405.2	10.5
LL24 (EF)	0.00565	2.407	0.048	3.477E-05	5.936E-07	1.69	399.7	10.5
LL16 (BS)	0.00452	1.926	0.039	3.139E-05	5.228E-07	3.94	445.0	11.6

BaS: Ballachulish Slate, LS: Leven Schist, EF: Eilde Flags, BS: Binnein Schist

probably situated below the bottom thrust boundary of the Barrovian metamorphic belt.

The K–Ar ages obtained from the Ballachulish Slate and the Leven Schist are younger than those of lower Formation of the Binnein Schist and apparently coeval with the Eilde Flags age. It seems that these formations, as well as the Eilde Flags, might also be on the lower side of the bottom boundary. However, it has also been argued that the Ballachulish Slate and the Leven Schist stratigraphically overlie the Binnein Schist (e.g. Treagus, 1974; Roberts 1976). Thus, other possible explanations cannot be ruled out to understand the relations between the K–Ar ages and the stratigraphy.

Summary

We analyzed phengites for their K–Ar ages from Barrovian metapelites in Loch Leven, Scotland. These data indicate a significant age-gap between ca. 445 Ma and ca. 400 Ma in the Loch Leven area. Integration of our data with previous geological and geochronological information from relevant metamorphic rocks suggests that the lower boundary of the Barrovian metamorphic belt is present as the Fort William Slide in the Loch Leven area.

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