

## Chromian illite-ankerite-quartz parageneses from the Kintail district of southern Ross-shire, Scotland

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### ABSTRACT

The Coire Dhuinnid fault zone contains emerald green chromian illite-ankerite-quartz rocks that are similar in appearance to the fuchsite(or mariposite)-carbonate-quartz parageneses that are commonplace in Archaean greenstone belts but which are rather rare in Phanerozoic rocks. The chromian illite contains 2.3 wt.% Cr<sub>2</sub>O<sub>3</sub>, low K<sub>2</sub>O (7.1–7.6 wt.%) and high H<sub>2</sub>O+ (5.7 wt.%), and it is a 1M polytype with ≤10% of an illite/smectite interstratification indicative of a formation temperature of *c.* 175–200°C. The host rocks contain high concentrations of Ni and Cr, and show low concentrations of Ti, Nb, Y and Zr, suggesting a former primitive mafic protolith (boninitic magma?); they are considered to be retrograde remnants of Lewisian rocks. The latter, and the associated rocks of the Moine Series, have been affected by CO<sub>2</sub> metasomatism that was accompanied by the addition of Ca(+Sr), Fe and Mg, and by the removal of Na from, and the addition of H<sub>2</sub>O to the Moine metasediments. Radiogenic isotope studies of mineral separates and whole rock from sample no. 43 yielded ages of 483 ± 2 Ma (Ar-Ar dating on Cr illite), 413 ± 12 Ma (K-Ar dating on Cr illite), and 322 ± 9 Ma (Rb-Sr dating on minerals and whole rock); the significance of this discrepant pattern is discussed.

**KEYWORDS:** Scottish Highlands, faulting, chromian illite, radiogenic isotopes, CO<sub>2</sub> metasomatism.

### Introduction

LOW-TEMPERATURE brittle deformational events in the Kintail district of the Northern Highlands of Scotland include the development of the Coire Dhuinnid fault zone that occupies a WNW-trending gorge in the upper reaches of the stream An Leth Allt (Fig. 1). The actual fault zone is 2–3 m in width but the mineralogical and textural effects of fault movement are observable in the country-rocks for distances up to 6 m on either side of the zone. The special interest in that locality is the occurrence of an emerald green chromian illite-ankerite-quartz rock (cf. listwanite; Kashkai, 1965). This is only the second recorded occurrence of a chromian mica in the Northern Highlands of Scotland; the first was from the Lewisian Gneiss Complex at Glenelg, immediately west of Loch Duich (see Fig. 1),

where fuchsite occurs with garnet, perhaps “as an alteration...of...eclogite in the omphacite of which chromium was present” (Peach *et al.*, 1910, p. 35). Moreover, the Coire Dhuinnid chromian illite-ankerite-quartz paragenesis is of particular interest because: (i) it is the first record of this rock type in Scotland (Clifford, 1959); (ii) it reflects low-temperature, fault-related CO<sub>2</sub> metasomatism that post-dated the regional Caledonian deformation and metamorphism; and (iii) elsewhere in the world similar parageneses, bearing fuchsite (chromian muscovite) or mariposite (chromian phengite), are often associated with gold deposits. This paper deals with the mineralogy, geochemistry and petrogenesis of the Scottish chromian illite-bearing parageneses and the associated rock types. In addition, the chemical and the Ar-Ar, K-Ar and Rb-Sr isotopic compositions of the chromian illite are presented.

**Regional setting**

The Northern Highlands are dominated by sedimentary rocks of the Moine Supergroup of Upper Riphean (*c.* 850–1000 Ma) age and, within this terrane, numerous inliers and tectonic slices of older (Archaean) Lewisian gneisses have been recognised (Holdsworth *et al.*, 1994). While there is some evidence of Grenville-age events (Holdsworth *et al.*, 1994, p. 24), the structure of the region is dominated by Caledonian deformation that has been described in terms of an earlier phase of regional ductile detachment such as the Sgùrr Beag thrust (Tanner, 1971; Barr *et al.*, 1986), and later large-scale open-to-tight upright folds (Harris and Johnson, 1991, p. 93). Deformation was accompanied by regional

metamorphism up to sillimanite grade (Kennedy, 1949; Fettes *et al.*, 1985). It has been suggested that the peak of the metamorphism was *c.* 455 Ma ago (Rogers and Pankhurst, 1993, p. 453), and that regional pegmatite emplacement followed shortly thereafter  $445 \pm 10$  Ma ago (van Breemen *et al.*, 1974). The Caledonian orogeny effectively ended with the intrusion of, *inter alia*, the Strontian granite, and the Ratagain granite complex at Glenelg (Fig. 1 (inset)),  $425 \pm 3$  Ma ago (Rogers and Dunning, 1991). Hutton and McErlean (1991) have recognized that sinistral strike-slip movement on the Strathconon fault (Fig. 1) took place during the Mid-Silurian crystallization of the main magmatic phases of the latter complex, and again during Early Devonian times (395–410 Ma ago).

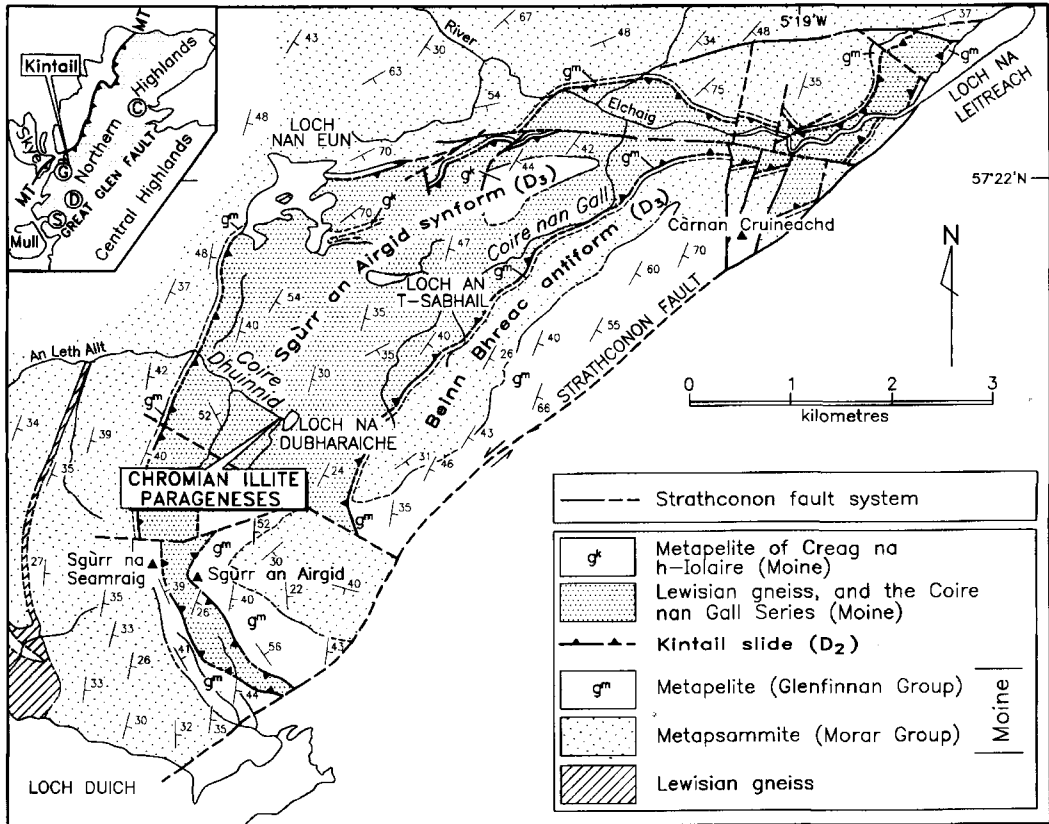


FIG. 1. The locality of the Coire Dhuinnid chromian illite parageneses shown in relation to the geology of part of the Kintail district of southern Ross-shire [after Clifford (1957a) with modifications in the west after May *et al.* (1993, p. 22)]. Inset: the Northern Highlands – abbreviations: C, Carn Chuinneag; D, Glen Dessary; G, Glenelg; MT, Moine Thrust; and S, Strontian.

The Kintail district of southern Ross-shire (see Fig. 1) is largely underlain by high-grade metamorphic rocks of the Lewisian Gneiss Complex, and the metapsammites (Morar Group) and metapelites (Glenfinnan Group) of the Moine Supergroup (Clifford, 1957a; May *et al.*, 1993). The structure of the area is characterised by the development of the Kintail slide and by subsequent isoclinal folding with the formation the Sgùrr an Airgid synform and the complementary Beinn Bhreac antiform (cf. Fleuty, 1974; May *et al.*, 1993, pp. 33–5). Ductile deformation was accompanied and outlasted by regional metamorphism with the development of kyanite-garnet ( $\pm$  staurolite  $\pm$  sillimanite) parageneses (Clifford, 1961).

The Coire Dhuinnid fault zone (Fig. 2a), formerly the An Leth Allt fault zone (Clifford, 1959), post-dated the regional metamorphism in the Kintail district, and is parallel or subparallel to faults to the southwest and in Glen Elchaig to the north (see Fig. 1) that are related to the Strathconon strike-slip fault. The fault zone is particularly well developed some 60 m downstream from the outflow of Loch na Dubharaiche (Fig. 1) where it is occupied by psammities and semipelites, together with concordant layers of emerald green chromian illite-ankerite-quartz parageneses (see Fig. 2a) that are the subjects of this paper. Clifford (1957a) originally grouped all of these rocks with the Coire nan Gall Series of Moine age.

### Petrology

The petrology of the various rock types that occur in association with the Coire Dhuinnid fault zone is dealt with under two subheadings, namely 'psammities and semipelites' and 'chromian illite parageneses'; all of the samples discussed in this work are now archived in the reference collections of the British Geological Survey in Edinburgh [see Appendix 1]. In each case the effects of hydration-CO<sub>2</sub> metasomatism are described in terms of three zones (Fig. 2b) of which only Zone C is clearly defined in the field. The northern boundary between Zones A and B has been drawn on the presence of fresh plagioclase in psammities and semipelites in the former, and its complete replacement by kaolinite in similar rocks in Zones B and C. In the absence of good outcrop, the southern boundary between Zones A and B has been arbitrarily drawn on Fig. 2b. Ankerite is characteristic of all litholo-

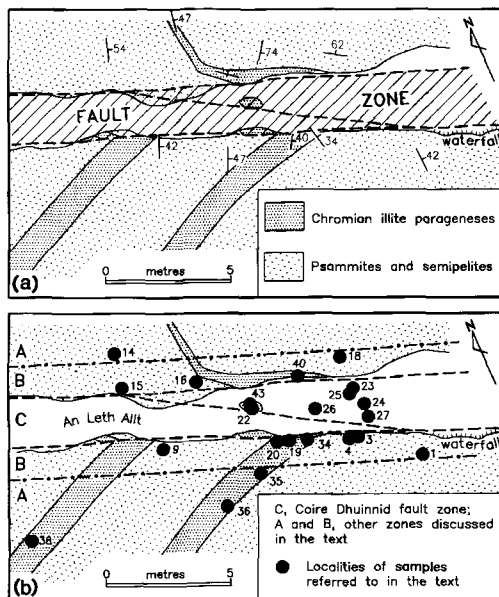


FIG. 2(a) Geological sketch map of the chromian illite occurrence in the upper part of Coire Dhuinnid (see Fig. 1) showing the strike and dip of the foliation; (b) localities of samples referred to in the text.

gies in the area, but its extent to the north and south is obscured by peat.

### Psammities and semipelites

These rocks are present in all three zones, and an example (sample no. 14) in Zone A consists of a granoblastic mosaic of oligoclase (An<sub>24</sub>), quartz and biotite; it differs from typical rocks elsewhere in the Sgùrr an Airgid synform in that it exhibits local cataclastic texture and contains microscopic veins of red brown-weathering ankerite. Equally, in Zone B, which is about 1 m wide, these rock-types (e.g. nos. 4, 9, 15, 16, 18; Fig. 2b) have largely retained their original metamorphic texture except that they are characterized by deformed quartz which shows intense strain-shadowing and arrays of sub-basal brittle fractures (Fig. 3a), by the complete pseudomorphous replacement of oligoclase by kaolinite, and by the presence up to 20% of ankerite. In addition, pale green muscovite occurs with chlorite after original biotite, and occasional large, fresh, remnant perthite crystals are also present (no. 15). Finally, narrow (<5 cm) ankerite veins (nos.

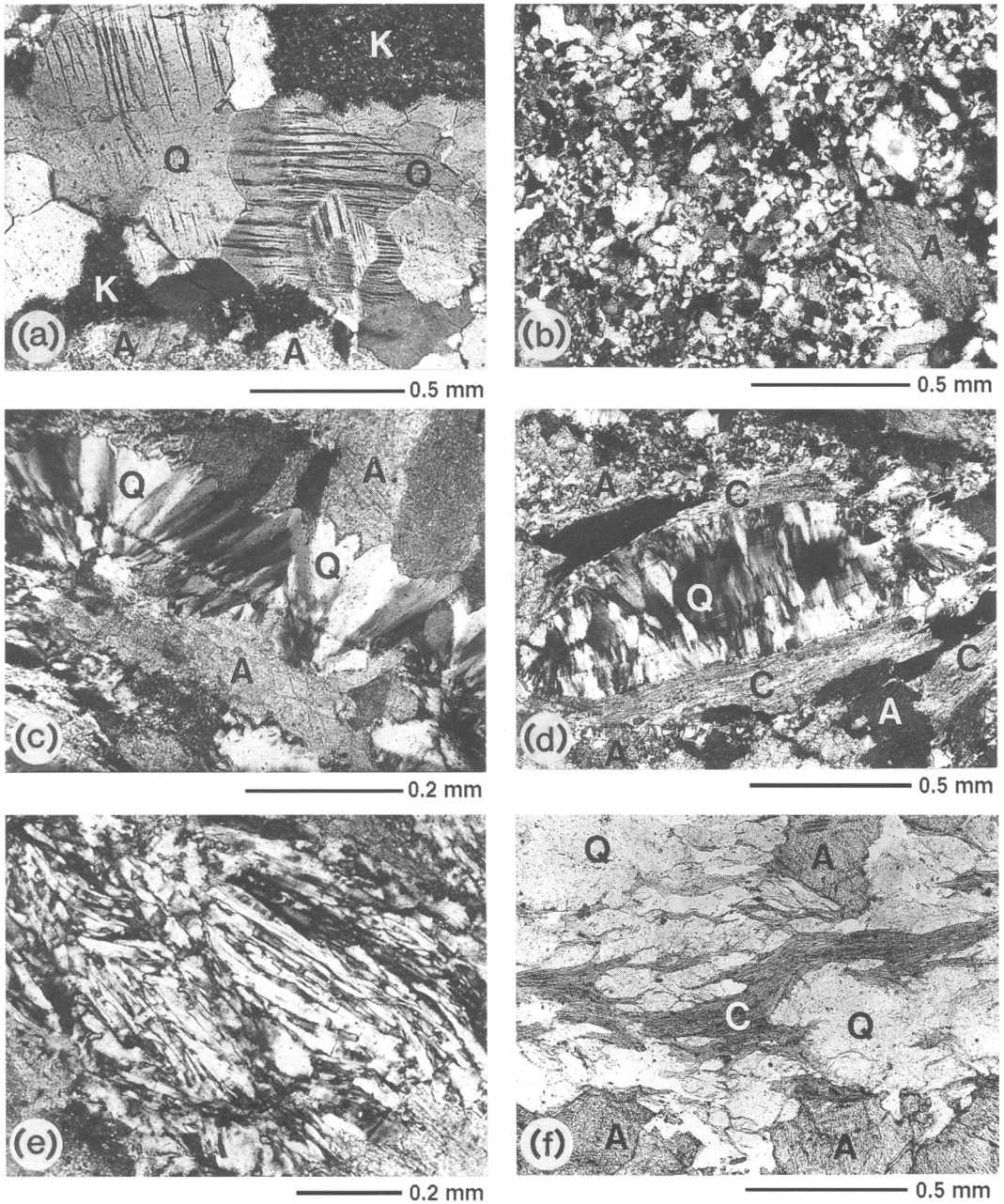


FIG. 3. Photomicrographs of rocks from Coire Dhuinid (abbreviations: A, ankerite; C, chromian illite; K, kaolinite; and Q, quartz): (a) psammite (Zone B, sample no. 16) composed of quartz showing arrays of sub-basal brittle-fractures, kaolinite pseudomorphs after plagioclase, and ankerite; (b) psammite (Zone C, no. 25) largely composed of intensely granulitized quartz with some later ankerite; (c)–(f) chromian illite-ankerite-quartz rocks showing (c) radiating euhedral quartz (Zone B, no. 19), (d) 'sharks-teeth' quartz with growth zoning (Zone C, no. 43), (e) 'sliver' quartz (Zone C, no. 22), and (f) a typical field of view of the chromian illite-ankerite-quartz rock (no. 43) from Zone C. Figs. 3a–e, crossed nicols; Fig. 3f ordinary light.

## CHROMIAN ILLITE-ANKERITE-QUARTZ PARAGENESES

1,3) are common in Zone B and consist of up to 50% carbonate, and fine-grained granulitized quartz. The partial chemical analysis of one such vein (Table 1, sample no. 3) shows the ankerite to be  $(\text{Ca}_{0.52}\text{Mg}_{0.35}\text{Fe}_{0.17})_{1.04}(\text{C}_{0.98}\text{O}_3)$ .

In Zone C the psammites and semipelites are identical to those of Zone B except that up to 90% of the quartz is present as fine-grained granulitized mosaics in certain samples (nos. 24–27; see Fig. 3*b*); locally, however, there are remnants of typical psammite (no. 23) with less than 10% of finely-granulated quartz. Pale green muscovite is ubiquitous, while later carbonate locally occurs as <1 cm-wide veins of ankerite (no. 26) or calcite (nos. 23, 25, 27).

The chemistry of representative samples of psammites and semipelites, and of the associated vein rocks is shown in Table 2 [analytical methods are given in Appendix 2]. The most pristine composition is that of sample no. 14 in which the oligoclase is still unaltered, and this is reflected by its  $\text{Na}_2\text{O}$  content of 4.17 wt.%. In contrast, all psammites and semipelites in Zones B and C (nos. 9, 15, 18, 23, 27) show that a dramatic loss of Na (to <0.08 wt.%  $\text{Na}_2\text{O}$ ) and an increase in  $\text{H}_2\text{O}^+$  contents up to 6.3 wt.%, accompanied the low-temperature kaolinization of the feldspar (cf. Thompson, 1970); concomitant with these changes all of the samples show a drop in  $\text{Al}_2\text{O}_3$ . Moreover, the psammites and semipelites in all zones contain carbonate that is reflected by the  $\text{CO}_2$  contents of 3.6 to 7.9 wt.% in these rocks in Zones B and C. The associated replacement veins (nos. 3, 26) show  $\text{CO}_2$  contents of >20% and, since these are accompanied by high Ca, Mg and Fe (Table 2, anals. 7 and 8), we conclude that a major part of these constituents and all of the  $\text{CO}_2$  in the psammites and semipelites have been introduced into the system during faulting.

#### Chromian illite parageneses

While these rocks are all composed of quartz, ankerite and micas, a dense green-brown colour is typical of examples from Zone A (nos. 35, 36, 38) whereas similar rocks from Zones B (nos. 19, 20, 34, 40) and C (nos. 22, 43) generally have a distinctive emerald green colour. The chemical compositions of these rocks are given in Table 2 (anals. 9–15); their interpretation is discussed on page 48.

Within Zone A, quartz occurs either as discrete grains showing strain-shadows and parallel

TABLE 1. Compositions of ankerites<sup>1</sup> from Coire Dhuinnid

Sample number	3	34	22
Zone	B	B	C
FeO*	6.60	4.22	5.05
MgO	7.59	7.70	8.67
MnO	0.13	0.18	0.18
CaO	15.86	14.37	16.22
CO <sub>2</sub>	23.89	21.82	24.58

Analysts, Joy R. Baldwin and Margaret H. Kerr.

<sup>1</sup>Chemical composition of the soluble (in 50% HCl) portions of: sample no. 3, replacement ankerite vein; and samples nos. 22 and 34, chromian illite-ankerite-quartz parageneses (see Fig. 2*b* for localities).

\*Total Fe as FeO.

fractures, or as lenses of radiating euhedral crystals. These two types also occur in Zone B; the former characterizes no. 34, while the latter predominates in sample no. 19 (Fig. 3*c*). In addition, close to the margin of the Coire Dhuinnid fault zone the quartz occurs as a granulitized fine-grained mosaic (nos. 20, 40). Finally, in Zone C (nos. 22, 43) the dominant quartz morphology is characterized by radiating crystals forming lenses that are elongated parallel to the rock foliation (Fig. 3*d*). In addition, an earlier phase of deformed ('sliver') quartz occurs as clusters of crystals (Fig. 3*e*) up to 1 mm in length and less than 0.03 mm in width oriented parallel to the rock foliation.

Ankerite is the most abundant mineral in the chromian mica parageneses in all three zones (A–C, Fig. 2*b*). In sample no. 38 in Zone A it occurs as large skeletal porphyroblasts while in samples nos. 35 and 36 it forms either aggregates of xenomorphic crystals, or lenses of sheaf-like crystals. In Zone B, in addition to these latter two types, an early phase of deformed and granulitized ankerite is present, while in Zone C the ankerite often shows intense deformation twinning and strain-shadowing. Partial chemical analyses of carbonates from two chromian illite-bearing parageneses (nos. 22, 34) are given in Table 1; both are ankerites  $(\text{Ca}_{0.52}\text{Mg}_{0.38}\text{Fe}_{0.12})_{1.02}(\text{C}_{0.99}\text{O}_3)$  that are somewhat less ferruginous than the vein carbonate (Table 1, sample no. 3).

Mica defines the irregular foliation in all of these rocks. Farthest from the fault, the biotite-

TABLE 2. Major (oxide wt.%) and trace (ppm) element concentrations in psammites and semipelites, vein rocks and chromian illite parageneses from Coire Dhuinnid; see Fig. 2*b* for localities

Sample number	psammites* and semipelites			veins			chromian illite parageneses									
	A	B	C	A	B	C	A	A	A	A	A	B	B	B	C	C
1	2	3	4	5	6	7	8	9	10	11	12	13	14	15		
14	18*	15	9	27*	23*	3	26	38	36	35	34	40	43	22		
Zone	A	B	B	B	C	C	B	C	A	A	A	B	B	C	C	C
SiO <sub>2</sub>	59.97	65.35	60.62	51.64	74.14	67.52	44.74	40.48	41.44	33.71	42.68	32.64	45.01	39.33	37.14	
TiO <sub>2</sub>	0.46	0.33	0.41	0.52	0.21	0.19	0.16	0.24	0.21	0.22	0.26	0.20	0.21	0.17	0.17	
Al <sub>2</sub> O <sub>3</sub>	17.16	14.32	15.59	15.38	5.86	10.08	2.47	5.49	5.89	4.94	7.01	4.84	5.54	3.52	2.95	
Fe <sub>2</sub> O <sub>3</sub>	1.3	0.06	0.00	0.21	0.04	0.02	0.6	0.0	0.68	0.7	0.7	0.7	0.50	0.4	0.20	
FeO	3.0	2.60	3.89	5.07	1.69	2.85	6.1	6.0	4.89	4.9	4.1	5.3	4.24	3.7	4.22	
MgO	2.37	1.99	2.10	3.96	1.56	2.54	6.99	6.89	7.89	9.16	7.89	9.41	7.89	9.74	9.50	
CaO	5.47	4.33	3.88	6.59	6.96	5.58	14.60	15.65	14.31	17.62	13.47	17.34	14.09	16.82	17.56	
MnO	0.08	0.08	0.08	0.10	0.08	0.08	0.09	0.11	0.12	0.14	0.11	0.11	0.12	0.11	0.12	
Na <sub>2</sub> O	4.17	0.03	0.07	0.03	0.03	0.02	0.89	0.91	0.01	0.87	0.92	0.88	0.02	0.02	0.90	
K <sub>2</sub> O	1.84	0.98	1.81	0.80	1.11	0.78	0.56	0.53	0.89	0.93	1.05	1.26	1.18	0.59	0.72	
CO <sub>2</sub>	3.1	5.7	3.6	7.9	6.4	6.8	21.7	21.7	20.4	25.5	19.1	25.2	19.1	23.9	25.1	
H <sub>2</sub> O+	0.13	4.19	6.23	6.26	1.99	3.81	1.37	2.46	2.34	1.54	2.85	2.48	2.67	1.91	1.46	
H <sub>2</sub> O-	0.49	0.20	0.13	0.41	0.14	0.12	0.17	0.54	0.23	0.35	0.34	0.72	0.22	0.12	0.87	
P <sub>2</sub> O <sub>5</sub>	0.35	0.15	0.22	0.25	0.09	0.08	0.07	0.09	0.05	0.08	0.06	0.06	0.05	0.05	0.06	
Total	100.89	100.31	98.63	99.12	100.30	100.47	100.51	101.09	99.35	100.66	100.54	101.14	100.84	100.38	100.97	
Rb	62	27	41	32	40	25	23	21	50	47	49	54	52	29	37	
Sr	517	82	149	73	121	99	334	379	584	523	400	447	517	899	929	
Y	9	6	10	11	6	5	5	10	4	10	9	4	7	5	5	
Zr	89	112	113	137	39	66	18	64	27	27	27	12	26	13	31	
Nb	9	8	10	9	5	5	4	6	6	7	8	8	7	4	5	
Co	14	9	13	15	6	9	16	15	34	33	33	26	17	28	43	
Ni	58	10	41	63	20	25	50	80	295	205	500	224	153	352	217	
Zn	78	52	62	76	49	45	86	91	75	86	75	93	61	47	53	
V	101	49	78	100	34	38	45	76	79	107	83	74	72	36	35	
Cr	102	12	196	166	44	6	29	35	2251	1005	2340	1580	1843	2480	2555	
Ba	437	282	1289	71	62	76	124	47	72	94	68	68	82	62	75	

Analysts, Sharon Farrell and Susan Hall, except for: (i) CO<sub>2</sub> values by Shamrielah Davids who also confirmed the low Na<sub>2</sub>O% in samples nos. 38, 40 and 43 as 0.03%, 0.03% and 0.01% respectively; and (ii) FeO for samples nos. 3, 14, 26, 34, 35, 36 and 43 by B & B Laboratories, Johannesburg. Bi, Cu and Sb are below the detection limit (10ppm) in all samples except for sample no. 9 which contains 10ppm Cu; Au < 1ppm in all samples.

like mica in sample no. 38 is almost opaque while elsewhere in Zone A (nos. 35, 36), and in Zone B (nos. 19, 20, 40), emerald green chromian illite is interlayered with greenish-brown biotite, except in sample no. 34 in which the only mica is chromian illite. This progressive change to chromian illite through Zones A and B is completed in Zone C in which the mica is entirely chromian illite that is particularly well-developed at localities nos. 22 and 43 where it occurs along the boundaries of the 'sharks-teeth' quartz aggregates (Fig. 3*d*); it clearly suffered deformation (Fig. 3*f*) that was associated with the growth of this radial quartz.

A typical modal analysis of chromian mica rock (no. 43) from Zone C is: ankerite, 54%; quartz, 34%; chromian illite, 11%. In addition, accessory minerals make up about 1% and include pyrite with inclusions of chalcopyrite (no. 22), clinocllore occurring with calcite in veins that cross-cut the foliation (no. 43), and zircon.

## Mineralogy

### *Chromian illite*

The XRD pattern for a chromian illite concentrate from sample no. 43 is shown in Fig. 4; in addition to illite, the pattern shows quartz, ankerite, chlorite and a trace of calcite. The observed maxima for the illite are 9.95, 4.98, 4.49 and 3.34 Å. Moreover, whilst the 3.62 and 3.06 Å reflections are weak using Cu radiation (see Fig. 4), they are obvious on the trace, recorded on paper, using X-rays generated from a Co target. The latter confirms that the chromian illite is a 1M mica polytype (Yoder and Eugster, 1955, p. 227) suggesting a minimum temperature of formation of about 175°C (Walker and Thompson, 1990, p. 317). Lines diagnostic of other polytypes are absent.

The (001) peak of the air dried chromian illite has a broad half-height peak width that becomes narrower after ethylene glycol solvation (Fig. 4).

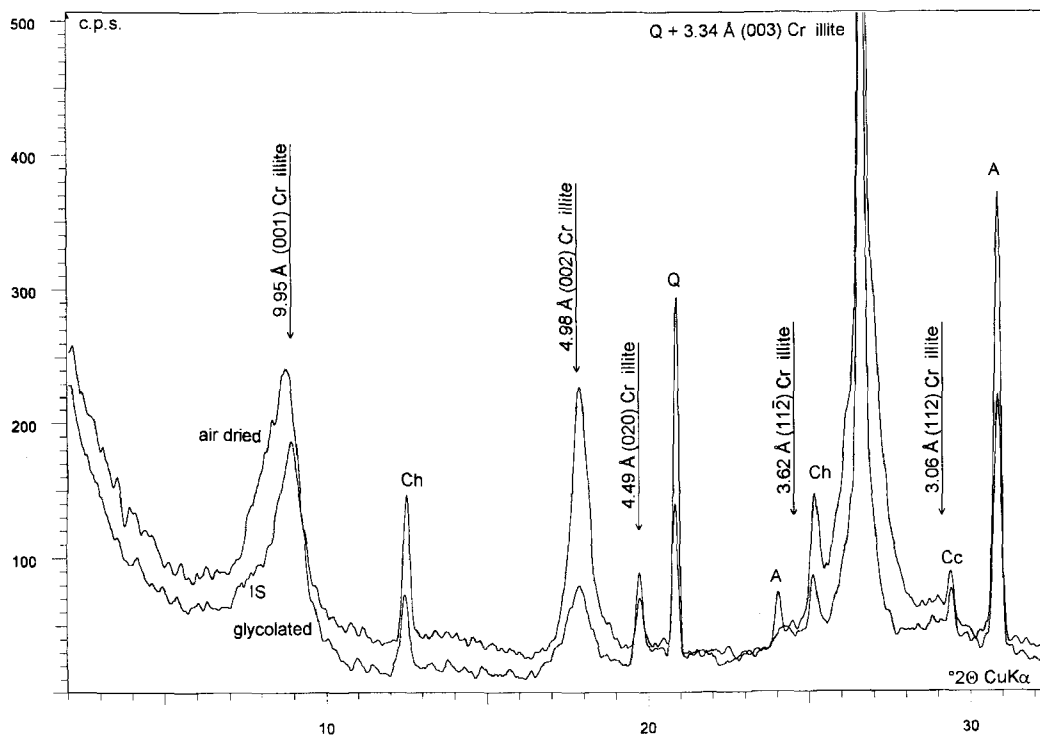


FIG. 4. X-ray powder diffraction patterns for air-dried and glycolated chromian illite concentrate from sample no. 43 from Coire Dhuinnid (abbreviations: A, ankerite; Cc, calcite; Ch, chlorite; c.p.s., counts per second; IS, illite/smectite interstratification; and Q, quartz).

Moreover, the trace after glycolation shows a distinct hump (IS) on the high Å side of the (001) illite peak that is indicative of an illite/smectite interstratification with an I:S ratio of about 10:1 (cf. Środoń, 1984, p. 341; Jennings and Thompson, 1986, p. 91; Walker and Thompson, 1990, p. 319). This implies a maximum temperature of formation of about 210°C, the temperature at which the percentage of illite in I/S increases to 100% (Jennings and Thompson, 1986).

The illite crystallinity (IC) of the 9.95 Å peak (cf. Kisch, 1990), based on the half-height peak width of glycolated sample no. 43, is  $0.74^\circ\Delta 2\theta$  compared to that of pegmatitic muscovite of  $0.085^\circ\Delta 2\theta$  achieved under the following experimental settings: scan speed  $0.5^\circ$  per minute; receiving slit width 0.05 mm; scan step size  $0.01^\circ 2\theta$ ; and digital recording system (cf. Kisch, 1991, p. 669). This high IC value is indicative of a very low crystallization temperature for the chromian illite (Merriman *et al.*, 1990, p. 37).

Chemical analyses of two chromian illites from Zone C in Coire Dhuinnid are presented in Table 3, one obtained by wet chemistry (anal. 1), the other by electron microprobe (anal. 2); except for  $K_2O$ , both analyses show identical major oxide ( $SiO_2$ ,  $Al_2O_3$ ,  $Cr_2O_3$ , total iron oxide and  $MgO$ ) contents. In Table 3 these new analyses are compared with that of 'illite chromifère' from the former Yugoslavia (anal. 3) and with published compositions of fuchsites (anal. 4–7) from U.S.A., Zimbabwe, New Zealand and Canada. Table 3 shows that, compared with the fuchsites, the Scottish chromian illite contains lower  $Al_2O_3$  and  $K_2O$  and higher total iron oxides,  $MgO$  and  $H_2O^+$ . Of these, the low  $K_2O$  and high  $H_2O^+$  contents (Table 3, anal. 1) are typical of illite (Deer *et al.*, 1962, p. 215; Newman and Brown, 1987, pp. 72–3) and, if the structural formula for the mica is calculated assuming that all of the  $H_2O^+$  represents hydroxyl groups, the interlayer (X) positions are only 75% filled (i.e.  $K+Ca+Na =$

TABLE 3. Compositions of chromian illites from Coire Dhuinnid and the former Yugoslavia compared with analyses of fuchsites from elsewhere in the world

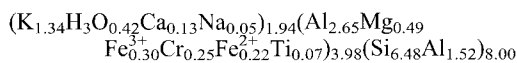
	1	2	3	4	5	6	7
	Chromian illites			Fuchsites			
$SiO_2$	47.40	47.65	50.05	44.4	44.7	46.35	46.26
$TiO_2$	0.76	0.22	0.40	2.1	0.50	0.28	0.0
$Al_2O_3$	25.75	25.97	22.83	31.7	34.0	29.69	26.50
$Cr_2O_3$	2.34	2.34	12.38	2.1	3.7	4.60	7.90
$Fe_2O_3$	2.96	6.24*	2.33	2.3	0.15*	0.23	0.69*
$FeO$	1.88		—	—		0.85	
$MnO$	0.01	—	—	0.05	n.d.	0.01	0.0
$MgO$	2.38	2.39	0.14	0.7	0.62	1.93	1.84
$CaO$	0.90	0.28	0.43	0.1	0.03	tr	0.0
$Na_2O$	0.17	0.04	0.24	1.1	0.96	0.78	0.0
$K_2O$	7.60	7.13	2.16	10.7	9.9	10.53	10.59
$H_2O^+$	5.73	n.d.	9.43	4.7	n.d.	4.69	n.d.
$H_2O^-$	2.22	n.d.	n.d.	n.d.	n.d.	0.12	n.d.
Total	100.10	92.26	100.34	99.95	94.56	100.31	94.56

Localities: (1) no. 22 (see Fig. 2b), Coire Dhuinnid - analyst, Erna Padget; (2) no. 43 (see Fig. 2b), Coire Dhuinnid - analyst, Anton P. le Roex; (3) Avala, Yugoslavia (Stangatchilovitch, 1956, p. 146) - X-ray data indicate the presence of a small amount of a kaolinitic mineral; (4) Acworth Township, New Hampshire, U.S.A. (Clifford, 1957b, p. 567); (5) O'Briens Claims, Zimbabwe (Schreyer *et al.*, 1981, p. 197); (6) Dead Horse Creek, Lake Wakatipu Region, western Otago, New Zealand (Hutton, 1942, p. 54) - analysis total includes  $BaO$  (0.15%),  $S$  (0.05%),  $F$  (0.04%) and  $P_2O_5$  (0.01%); (7) Baie Verte, Newfoundland (Chao *et al.*, 1986, p. 723) - analysis total includes  $NiO$ , 0.78%.

\*Total Fe as  $FeO$ ; n.d. = not determined.



1.5). The major deficiency of potassium and the excess of water most likely reflects the presence of  $H_3O^+$  (oxonium) cations in the interlayer positions (Brown and Norrish, 1952). With this assumption, the structural formula for the chromian illite, based on 20(O) and 4(OH) per structural unit, is:



However, this illite is associated with  $\leq 10\%$  of an illite/smectite interstratification (see Fig. 4), and the very low K-concentrations that characterize the members of the smectite group ( $K_2O$  generally  $< 0.1\%$ ; Newman and Brown, 1987, pp. 49–57) imply that the  $K_2O$  content of the chromian illite is higher than the measured value (cf. Table 3, anal. 1). This requires a modest increase in the number of K-cations in the above idealized formula, but other adjustments are uncertain because of the wide compositional range of smectites (Newman and Brown, 1987).

#### Chromian sodian mica

The strong green-brown rock colour in certain chromian illite parageneses in Zone A reflects the presence of almost opaque biotite-like micas. As a result of the high Cr content of c. 2200 ppm in the rock (see Table 2), examples of these micas have been analysed by EMP from sample no. 38 furthest from the Coire Dhuinnid fault zone (Fig. 2b). They have a wide variation in their chemistry but the most uniform composition (Table 4, anal. 1) shows high  $Cr_2O_3$  and  $Na_2O$  contents of 2.39 wt.% and 2.43 wt.% respectively. The calculated structural formula [ $X_{2.06}Y_{4.26}Z_8$  for 22(O)] for this mica shows a 6% excess in the Y site-occupancy that presumably reflects some inhomogeneity in the analysed material. The Na-content suggests  $>30\%$  of the paragonite component, while  $Cr+Fe+Mg+Ti$  constitute  $>40\%$  of the Y sites and may explain the dense colour. However, the low  $Na_2O$  content of the rock (0.01–0.03 wt.%; Table 2) indicates that mica of this specific composition makes up only about 1% of the modal mineralogy of the rock. Other micas in this paragenesis (Table 4, anals. 2 and 3) contain higher  $SiO_2$  (50 wt.% and 56 wt.%), greatly reduced  $Na_2O$  (1.1 wt.% and 0.01 wt.%) and lower  $Cr_2O_3$  (1.6 wt.% and 1.4 wt.%), and show major deficiencies in the X site-occupancy ( $\Sigma$ interlayer: 1.62 and 1.20).

TABLE 4. Compositions of chromian micas from sample no. 38 from Coire Dhuinnid

	1	2	3
$SiO_2$	47.26	50.12	56.48
$TiO_2$	0.11	0.13	0.01
$Al_2O_3$	23.17	23.77	22.65
$Cr_2O_3$	2.39	1.55	1.38
$FeO^*$	7.53	7.47	4.61
MnO	0.01	0.01	0.01
MgO	2.95	3.58	3.50
CaO	0.12	0.20	0.64
$Na_2O$	2.43	1.13	0.01
$K_2O$	7.75	7.49	6.72
Total	93.72	95.44	96.26

Analyst, Anton P. le Roex.

\*Total Fe as FeO.

These 'micas' are clearly inhomogeneous or are intergrowths.

#### Isotope data

Isotope studies of mineral separates from chromian illite-ankerite-quartz paragenesis no. 43 (see Fig. 2b) show a diverse pattern of Ar-Ar, K-Ar and Rb-Sr ages ranging from 480 Ma to 320 Ma [analytical methods are given in Appendix 2]. Of these, the  $^{40}Ar$ - $^{39}Ar$  data for the chromian illite (Table 5) give a total fusion age of  $498 \pm 2$  Ma, and the spectrum (Fig. 5) is made up of two phases: an initial

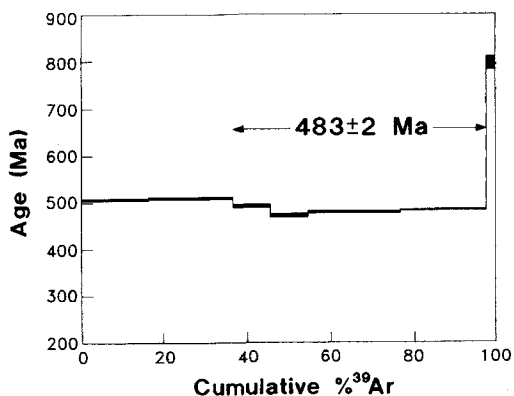


FIG. 5.  $^{40}Ar$ - $^{39}Ar$  spectrum for chromian illite (sample no. 43) from Coire Dhuinnid; see Table 5 for analytical data.

TABLE 5.  $^{40}\text{Ar}$ - $^{39}\text{Ar}$  data for chromian illite (sample no. 43) from Coire Dhuinnid

Temp °C	$^{39}\text{Ar}_K$ Vol.	$^{37}\text{Ar}_{Ca}$ $\times 10^{-9}$	$^{38}\text{Ar}_{Cl}$ $\text{cm}^3$	Ca/K	$^{40}\text{Ar}/^{39}\text{Ar}_K$	%Atm $^{40}\text{Ar}$	Age Ma	Error Ma	% $^{39}\text{Ar}_K$
550	1.41	0.08	0.05	0.12	126.7	2.4	506.7	1.5	17.0
660	1.60	1.10	0.03	1.36	127.2	7.4	508.7	1.3	19.4
715	0.75	0.34	0.01	0.91	123.4	2.1	495.2	2.3	9.1
790	0.80	0.04	0.02	0.10	117.4	6.0	474.1	2.6	9.7
895	1.80	0.05	0.03	0.06	119.0	5.8	479.7	0.9	21.7
1000	1.75	0.08	0.03	0.09	119.6	6.8	481.8	0.9	21.2
1320	0.16	0.51	0.01	6.18	214.8	24.0	790.7	13.4	2.0

Analyst, David C. Rex. Sample weight 0.01247g; weight %K 3.7;  $^{40}\text{Ar}$  (volume of radiogenic  $^{40}\text{Ar}$ )  $824 \times 10^{-7} \text{ cm}^3 \text{ g}^{-1}$ ; gas volumes corrected to STP; J value  $0.00256 \pm 0.5\%$  (calibration using hornblendes Hb3gr, MMHbl and Fy12a (see Roddick, 1983)); errors at  $1\sigma$ .

temperature phase up to 715°C, comprising 36.4% of the gas with an age of  $507 \pm 1$  Ma; and a high-temperature phase (steps 3–6) comprising 61.7% of the gas, with an age of  $483 \pm 2$  Ma. If these ages reflect the time of Coire Dhuinnid faulting, then the latter represents a minimum age for

ductile deformation/regional metamorphism in the Kintail district (Option 1, Fig. 6; cf. Dewey and Pankhurst, 1970). However, the chromian illite is characterized by comparatively high IC of 0.74 (see p. 44), which Dong *et al.* (1995) have shown to be typical of illites that have suffered

Northern Highlands (Harris & Johnson, 1991)	Kintail district of southern Ross-shire
Intrusion of post-tectonic granites (400–414 Ma)	$322 \pm 9 \text{ Ma}^5$ : Rb–Sr (chromian illite–clinocllore–whole rock) age, Coire Dhuinnid fault zone
Moine Thrust Zone. Intrusion of Loch Borrolan syenite ( $430 \pm 4 \text{ Ma}^3$ )	$413 \pm 12 \text{ Ma}^5$ : K–Ar (chromian illite) age, Coire Dhuinnid fault zone
Open-to-tight folds: axial planes N–S to NE–SW	Intrusion of Ratagain complex ( $425 \pm 3 \text{ Ma}^4$ )
Intrusion of Glen Dessary syenite ( $456 \pm 5 \text{ Ma}^2$ )	Option 2:
Interleaving of Lewisian basement along Sgùrr Beag and other thrusts	D <sub>3</sub> Tight folding; Sgùrr an Airgid and Beinn Bhreac folds
	D <sub>2</sub> Kintail slide
	$483 \pm 2 \text{ Ma}^5$ : $^{40}\text{Ar}$ – $^{39}\text{Ar}$ (chromian illite) age, Coire Dhuinnid fault zone
	Option 1:
	D <sub>3</sub> Tight folding; Sgùrr an Airgid and Beinn Bhreac folds
	D <sub>2</sub> Kintail slide
Intrusion of Carn Chuinneag granite ( $550 \pm 10 \text{ Ma}^1$ )	

FIG. 6. The sequence of Lower Palaeozoic events suggested for the Northern Highlands (Harris and Johnson, 1991) compared with those in the Kintail district of southern Ross-shire. Sources of geochronological data: <sup>1</sup>Long (1964) and Pidgeon and Johnson (1974); <sup>2</sup>van Breemen *et al.* (1979a); <sup>3</sup>van Breemen *et al.* (1979b); <sup>4</sup>Rogers and Dunning (1991); <sup>5</sup>this work.

## CHROMIAN ILLITE-ANKERITE-QUARTZ PARAGENESES

TABLE 6. Rb-Sr data for the chromian illite-ankerite-quartz paragenesis (sample no. 43) from Coire Dhuinnid

	Rb (ppm)	Sr (ppm)	$^{87}\text{Rb}/^{86}\text{Sr}$	$^{87}\text{Sr}/^{86}\text{Sr}$
Chromian illite (a) after leaching with 2.5 M HCl	261.25	7.94	99.81	$1.17174 \pm 14$
Leachate (a') from (a)	4.71	351.39	0.039	$0.70829 \pm 30$
Clinochlore (b)	373.49	99.85	10.88	$0.76566 \pm 3$
Chromian illite (+ankerite) (c)	158.65	43.40	10.65	$0.76871 \pm 3$
Whole-rock (d)	37.28	781.16	0.138	$0.71848 \pm 3$

Analyst, Rodney Green.

high levels of  $^{39}\text{Ar}$  recoil-loss during the irradiation of potassium in the loosely-held lattice sites in the illite structure (McDougall and Harrison, 1988; Reuter and Dallmeyer, 1989). We conclude, therefore, that the Ar-Ar age may be geologically meaningless.

The K-Ar and Rb-Sr ages for the chromian illite are both consistent with the timing of the structural/metamorphic events established elsewhere in the Northern Highlands (Fig. 6), notably the syntectonic intrusion of the Glen Dessary syenite  $456 \pm 5$  Ma ago (van Breemen *et al.*, 1979a; Roberts *et al.*, 1984) and amphibolite facies regional metamorphism/pegmatite emplacement that continued until 430 Ma ago (van Breemen *et al.*, 1979a). However, the Rb-Sr age (Fig. 7) of  $322 \pm 9$  Ma (MSWD, 18;  $1_{\text{Sr}}$ ,

$0.71785 \pm 0.00009$ ) results from the omission of the leachate data (a', see Table 6); it is, moreover, essentially a two-point isochron age in which the chromian illite fraction (a) has such a high Rb/Sr that it controls the age. If all five data points (Table 6) are included in the regression the resulting age of  $347 \pm 100$  Ma (MSWD, 1593;  $1_{\text{Sr}}$ ,  $0.71297 \pm 0.00171$ ) reflects a considerable scatter of data points suggesting that the Rb-Sr system has been heavily disturbed. Without corroborating data, therefore, it is imprudent to ascribe a geological meaning to these ages.

Finally, while the K-Ar isotope data may reflect the effects of argon loss and/or the presence of excess  $^{40}\text{Ar}$  (Kelley, 1988), it is equally possible that the age of  $413 \pm 12$  Ma (Table 7) dates the brittle deformation and fluid movement associated with the Coire Dhuinnid fault. The latter interpretation is consistent with the inference (see p. 39) that the fault is a minor expression of major faulting such as the Great Glen fault and the Strathconon fault (see Fig. 1) whose strike-slip movement was initiated in Lower Silurian times, 420–430 Ma ago, and was largely completed in Early Devonian times, 390 Ma ago (Watson, 1984, pp. 200–2; Hutton and McErlean, 1991).

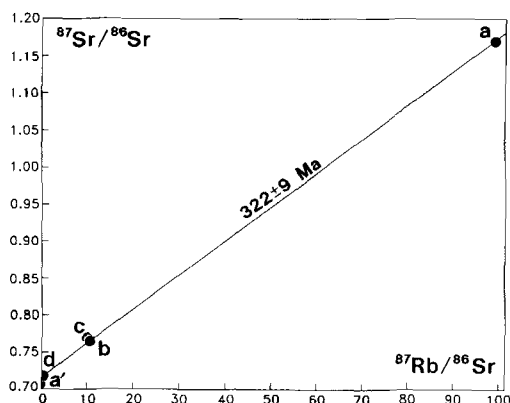


FIG. 7. Rb-Sr data for chromian illite-ankerite-quartz rock (sample no. 43) from Coire Dhuinnid; see Table 6 for the analytical data and the designation of a, a', b, c and d.

TABLE 7. K-Ar data for chromian illite (sample no. 43) from Coire Dhuinnid

%K	Vol $^{40}\text{Ar}$ rad scc/g $\times 10^{-5}$ STP	% $^{40}\text{Ar}$ rad	Age (Ma)
5.13	9.2591	95.9	$413 \pm 12$

Analyst, David C. Rex.

## Discussion and conclusions

Following the classic work of Whitmore *et al.* (1946) on parageneses bearing chromian micas (fuchsite and mariposite), Martyn and Johnson (1986) have summarized the associations and note that they include:

(1) *metamorphic parageneses* either (a) with corundum and/or aluminosilicates that are the result of greenschist-amphibolite facies metamorphism of the products of exhalative alteration of komatiitic lavas (Schreyer *et al.*, 1981; Martyn and Johnson, 1986); or (b) as the product of regional metamorphism of former (perhaps chromite-bearing) quartzite and quartz conglomerate (Frankel, 1939; Clifford, 1957b; Padget *in* Holmsen *et al.*, 1957; Leo *et al.*, 1965).

(2) *metasomatic parageneses* represented by fuchsite (or mariposite)-ankerite-quartz assemblages commonly associated with vein gold or antimony mineralization. These are the most common parageneses for chromian micas, and by far the majority (>90%) of examples occur in Archaean terranes, notably in Canada (Cooke, 1922), Western Australia (Stillwell, 1929; Sund *et al.*, 1984) and South Africa (Pearson and Viljoen, 1986). Important younger occurrences of these rock types are well known in the Sierra Nevada foothills of California, notably in the Mother Lode System (Knopf, 1929) and in the Alleghany District (Ferguson and Gannett, 1932); they have yielded K-Ar (mariposite) and Rb-Sr (mariposite-carbonate) ages of 115–120 Ma (Böhlke and Kistler, 1986).

The chromian illite-ankerite-quartz parageneses at Coire Dhuinnid in Scotland are clearly related to (2). In early work in the Archaean rocks of Western Australia, Simpson and Gibson (1912) and Feldtmann (1916, p. 38) suggested that the fuchsite-carbonate rocks were original basic/ultrabasic rocks. Simpson and Gibson (1912) proposed that the fuchsite was formed by the action of K-bearing waters on chromite and aluminium silicates, and that the original silicates suffered replacement by Ca, Mg and Fe carbonates. A similar relationship between K metasomatism and CO<sub>2</sub> metasomatism of ultrabasic rocks has been suggested for fuchsite-carbonate rocks in the Murchison Range in South Africa (Pearson and Viljoen, 1986, p. 306), and at the Kerr Addison Mine in Ontario (Downes, 1981, p. 69). Colvine *et al.* (1984) suggested that the fuchsite-ankerite-quartz rocks, which are major gold-ore host-rocks at the latter locality, represent

highly carbonatized and sericitized komatiitic flows.

In the Coire Dhuinnid chromian illite-ankerite-quartz parageneses, the Ca, (Fe+Mg), Sr and CO<sub>2</sub> contents (see Table 2, anal. 9–15) are identical to those of the ankerite veins (Table 2, anal. 7 and 8) of obvious replacement origin, thus supporting the view that a major part of these constituents has been added to the chromian illite system. A possible source-model for the CO<sub>2</sub> in similar associations in the Archaean in Canada has been suggested by Colvine *et al.* (1984, p. 62) who argued that mantle-derived basaltic liquid ascended into a granulite facies lower crust where it became saturated with CO<sub>2</sub> and H<sub>2</sub>O; magma and hydrothermal CO<sub>2</sub>+H<sub>2</sub>O fluids then rose towards the surface along faults. With this model, the Ca, Mg and Fe for ankerite formation were presumably scavenged from the deeper crust by these fluids.

In contrast, the low concentrations of high field strength elements such as Ti, Nb, Y and Zr, and the high Ni (153–500 ppm) and Cr (1005–2555 ppm) that characterize the chromian illite parageneses (see Table 2, anal. 9–15) are believed to be pristine and they suggest a former primitive mafic magma, perhaps of boninitic composition (cf. Crawford *et al.*, 1989, p. 9). Since such unusual compositions are foreign to the Moine Series, we conclude that the chromian illite rocks are carbonated and hydrated remnants of tight infolds of Lewisian mafic igneous rocks that are correlative with larger bodies of these rocks elsewhere in the core of the Sgurr an Airgid synform (Clifford, 1957a; May *et al.*, 1993, p. 34).

Isotope studies of chromian illite-bearing sample no. 43 from Coire Dhuinnid have yielded the following ages: 482 ± 3 Ma (Ar-Ar dating on Cr illite); 413 ± 12 Ma (K-Ar dating on Cr illite); and 322 ± 9 Ma (Rb-Sr dating on Cr illite-clinocllore-whole rock). This discordance is not surprising because the 1M nature of the chromian illite and the presence of ≤10% of interstratified illite-smectite indicate a crystallization temperature of *c.* 175–200°C (Walker and Thompson, 1990, p. 317; Jennings and Thompson, 1986, p. 92) that is well below the anchizone-epizone boundary (*T.* *c.* 280 ± 30°C (Bucher and Frey, 1994, p. 272)) near which K-Ar and Rb-Sr data and the Ar-Ar release spectra are concordant (Hunziker *et al.*, 1986). A very low temperature is also confirmed by the complete replacement of feldspar by kaolinite in Zones B and C, and by the high IC of 0.74 of the chromian

illite that indicates crystallization in the diagenetic zone [zeolite facies] (Merriman *et al.*, 1990, p. 37; Warr, 1996).

The Coire Dhuinnid faulting, with its associated H<sub>2</sub>O and CO<sub>2</sub> metasomatism and chromian illite formation, post-dates the regional Caledonian ductile deformation/metamorphism, and the K-Ar age of 413 ± 12 Ma of the mica may reflect the timing of those low-temperature events. This implied Palaeozoic age for the formation of the chromian illite-ankerite-quartz parageneses is unusual since, with the exception of Mesozoic examples of mariposite-bearing associations, the majority of similar rock types are Archaean. If, however, the pristine source of the Coire Dhuinnid chromian illite rocks was indeed Lewisian mafic rocks, as argued above, then their chromium-bearing precursors were of Archaean age. Finally, notwithstanding the clear identity of these Scottish parageneses with similar rocks in the gold-bearing terranes of Canada, Western Australia and California, the Au concentrations in all rocks from Coire Dhuinnid are less than 1ppm (Table 2) and are therefore unlikely to be economic.

### Acknowledgements

We are very grateful to the following: the Research Committee of the University of the Witwatersrand for financial support for this research project; Sharon Farrell, Susan Hall, Shamrielah Davids, Joy Baldwin and the late Margaret Kerr for rock analyses; Erna Padget for the analysis of chromian illite; the late Finlay Johnston for the photomicrographs; Diane du Toit for draughting the illustrations; Henia Czekanowski for photography; Pat King for secretarial services; Dion Brandt for U-stage determinations of quartz deformation lamellae; and Lew Ashwal, Jay Barton, Rudi Boer, Guy Charlesworth, Don Hunter, Don Mallick, Dave Phillips, Graeme Rogers, and the two journal reviewers R.J. Merriman and an anonymous referee, whose constructive comments resulted in real improvements in the presentation of our data. This paper is dedicated to the memory of Oleg von Knorring.

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## Appendices

### Appendix 1: Rock specimens

The rock samples from Coire Dhuinnid (TNC field location 1551) described in this paper are now archived in the reference collections of the Scottish Office of the British Geological Survey, Murchison House, West Mains Road, Edinburgh. The field numbers [and correlative BGS reference numbers (N262, etc.)] are as follows: 1, N262; 3, N263; 4, N264; 9, N265; 14, N266; 15, N267; 16, N268; 18, N269; 19A, N270A; 19B, N270B; 20, N271; 20L, N272; 22, N273; 22/0, N274; 22/1, N275; 22/2, N276; 23, N277; 24, N278; 25, N279; 26; N280; 27, N281; 34, N282; 35, N283; 36, N284; 38, N285; 40, N286; 42, N287; 43, N288.

### Appendix 2: Analytical techniques

The rock samples were analysed for major and trace elements using the Phillips PW1400 XRF spectrometer with a rhodium target X-ray tube. The Norrish and Hutton (1969) fusion technique was used for major elements, and pressed powder pellets with calculated mass absorption coefficient correction for matrix effects were used for trace elements and Na<sub>2</sub>O (Feather and Willis, 1975). FeO values were determined by titrimetry, while the CO<sub>2</sub> abundances were calculated following duplicate determinations of CaCO<sub>3</sub> using the Karbonat-bombe method of Birch (1981). H<sub>2</sub>O–

was determined gravimetrically, and H<sub>2</sub>O+ was obtained from the loss on ignition after correction for CO<sub>2</sub> and iron oxidation.

For the isotope studies, the chromian illite was separated by magnetic techniques, and the concentrate was washed in 5% acetic acid to remove extraneous carbonate; the low K value of 3.7% for the Cr illite used in the Ar-Ar analyses indicates that this process was incomplete. Details of the irradiation and the argon-extraction procedures are given by Rex *et al.* (1993). For the K-Ar determinations, the potassium content was determined by flame photometry using a Li-internal standard on a Corning-Eel 480 flame-photometer. Argon was measured by isotope dilution, while the ratios of the argon isotopes were measured on a modified MS10 mass spectrometer fitted with computer data acquisition following experimental details given in Rex (1994). Rb-Sr isotopic analyses were made on a VG Micromass 30 mass spectrometer; full analytical details are given by Cliff *et al.* (1985). Decay constants used in all of the above methods are from Steiger and Jäger (1977). Analytical uncertainties are for 95% confidence limits.

[Manuscript received 5 January 1998;  
revised 27 May 1998]