

The retrogression of ultramafic granulites from the Scourian of NW Scotland

JANE D. SILLS

Geology Department, University of Leicester, Leicester, LE1 7RH

ABSTRACT. Scourian ultramafic granulites from the Assynt region of Sutherland were retrogressed to amphibolite facies during the Inverian and in Laxfordian shear zones. Inverian assemblages are dominantly chlorite and tremolite with minor anthophyllite and dolomite. Laxfordian assemblages are cummingtonite, chlorite, and dolomite with only minor amounts of tremolite. Retrogression took place on a falling temperature path with temperatures between 750 and 550°C, and with H₂O forming more than 90% of the fluid phase. Initially retrogression was caused by the influx of large volumes of hydrous fluid associated with the development of Inverian folds. During the Laxfordian shearing P_{CO_2} was increased slightly. Serpentinization of olivine occurred at a much later stage.

THIS paper considers the retrogression of Scourian ultramafic granulites from the Assynt region of Sutherland during both the Inverian and Laxfordian events. The ultramafic granulites concerned form part of layered ultramafic-gabbro bodies (Bowes *et al.*, 1964) metamorphosed at granulite facies during the Scourian event at *c.* 2.7 b.y. (Chapman and Moorbath, 1977). The Lewisian gneisses of Assynt have suffered two further tectono-metamorphic events (Sheraton *et al.*, 1973; Tarney, 1973); the first of these, the Inverian, caused widespread retrogression to amphibolite facies associated with the development of north-west-trending monoclinical folds. Most of the retrogression occurred before the intrusion of the Scourie dykes. The gneisses were further affected by localized, discrete Laxfordian shear zones which caused a substantial reduction in grain size and the development of a new penetrative foliation and lineation. There are three types of retrogressive metamorphism affecting the ultramafic rocks:

- (i) Retrogression without complete recrystallization in or near Inverian fold belts.
- (ii) Recrystallization accompanied by shearing in Laxfordian shear zones.
- (iii) Serpentinization of olivine and orthopyroxene.

In this paper the mineralogical changes, the P - T

conditions, and the composition of the fluid phase that caused the retrogression and consequent chemical changes will be discussed.

Petrography

The ultramafic granulites have an equigranular, granoblastic texture typical of granulite-facies rocks (fig. 1a). They have the assemblage: orthopyroxene-clinopyroxene-olivine-pargasite-spinel-magnetite with harzburgite occurring locally. This assemblage is recognized as being entirely metamorphic with the normative mineralogy classifying the rocks as plagioclase-peridotites.

The first sign of retrogression is the exsolution of opaque oxides from hornblende and pyroxene along cleavage planes and grain boundaries, this effect being most pronounced in hornblende (fig. 1b). This is due to the exsolution of excess TiO₂ from granulite-facies pargasite and is associated with a change in colour from brown to green. When the rock has completely retrogressed, the Inverian assemblage is typically: tremolite-chlorite-magnetite ± anthophyllite ± dolomite ± ilmenite. In incompletely retrogressed assemblages anthophyllite may partially replace orthopyroxene and chlorite needles replace pargasite (fig. 1c). Tremolite occurs either as euhedral grains or as fine-grained intergrowths with chlorite and minor anthophyllite, these intergrowths pseudomorphing the granulite-facies texture. Colourless chlorite occurs as needles cutting hornblende or pyroxenes (fig. 1c), as intergrowths with tremolite and as rims around opaque oxides. Minor dolomite occurs but is more abundant in shear zones.

In Laxfordian shear zones a strong planar fabric develops with a parallel alignment of platy minerals and a segregation into carbonate and silicate layers. The following assemblage is typical: chlorite-anthophyllite or cummingtonite-dolomite-magnetite ± tremolite ± calcite ± hematite. Dolomite is the dominant carbonate mineral with only subsidiary amounts of fine-grained anhedral calcite. There is

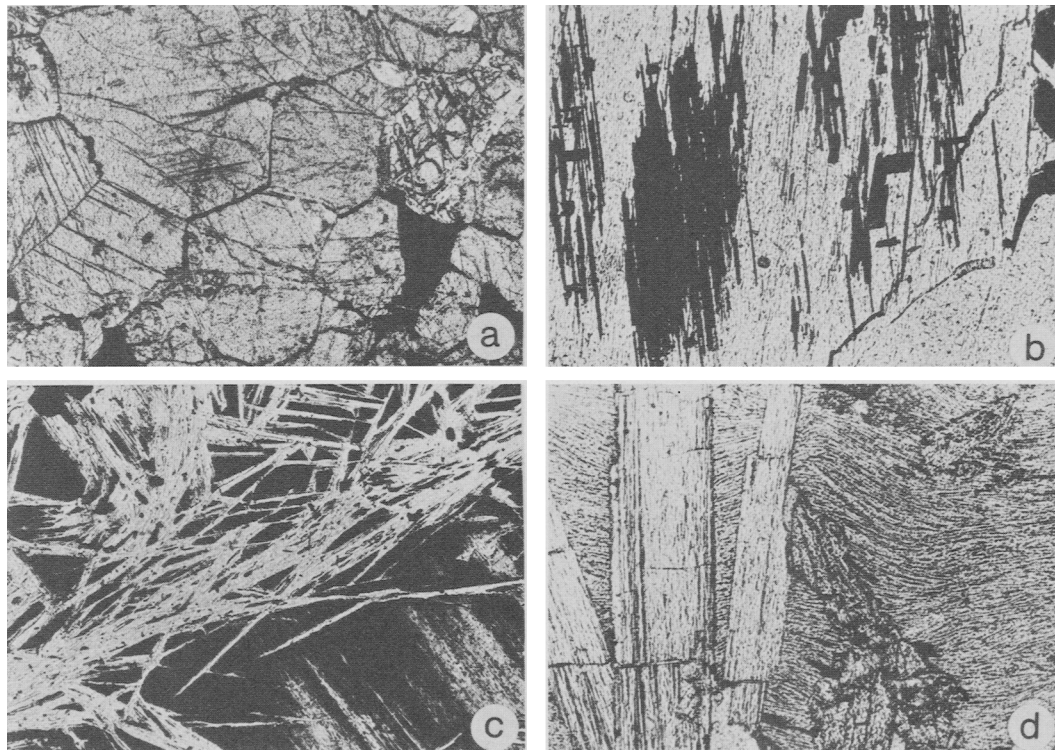


FIG. 1. (a) Granulite showing typical texture. Field of view 3.2 mm. Sample W1. (b) Exsolution of opaque oxides from granulite facies hornblende. Field of view 2.00 mm. Sample W49. (c) Chlorite needles cutting relict hornblende grain. Field of view 3.2 mm. Sample W9. (d) Cummingtonite laths cutting folded chlorite matrix, sample S2, from Laxfordian shear zone. Field of view 2.00 mm.

a considerable reduction in the amount of tremolite, which may disappear altogether to be replaced by a calcium-free amphibole and dolomite. The amphibole is cummingtonite as it has a small extinction angle. In the 0.5 km wide Canisp shear zone (Sheraton *et al.*, 1973; Sills, 1981) dolomite often aggregates in fold hinges leaving a rock composed dominantly of chlorite and cummingtonite. The folded schistose chlorite matrix is cut by post-deformational laths of cummingtonite (fig. 1d). This migration of dolomite thus leaves a rock with no calcium mineral.

The assemblages described above are plotted on fig. 2. The SiO_2 -CaO-MgO diagram (projected from Al_2O_3 - CO_2 - H_2O) gives a good representation of the retrogressed assemblages but is less good for the granulite-facies assemblage as Al_2O_3 is contained in pargasite, spinel, and pyroxene. From these diagrams it is possible to write a series of reactions (Table I): reactions 1 to 6 accounting for the formation of the Inverian tremolite-chlorite \pm anthophyllite assemblages and reactions 7 to 9 accounting for the near disappearance of tremolite

to form dolomite-cummingtonite assemblages. Dolomite producing reactions (such as reaction (7); Table I) release excess silica which does not appear as quartz. In the Canisp shear zone, there are abundant quartz veins especially in the vicinity of ultramafic rocks. These veins are folded and therefore predate the later phases of deformation.

All the granulite-facies samples show some serpentinization of olivine and occasional completely serpentinized rocks are found. Serpentinites have minor chlorite and abundant magnetite. The age of serpentinization is not clear, but Tarney (1973) demonstrated that it post-dated the Inverian and Laxfordian metamorphism of picritic Scourie dykes. The amount of serpentinization appears to increase eastwards across the Lewisian outcrop towards the Moine thrust and could possibly be of Caledonian age.

Mineral chemistry

Minerals were analysed using a Cambridge Instruments Microscan V electron microprobe at

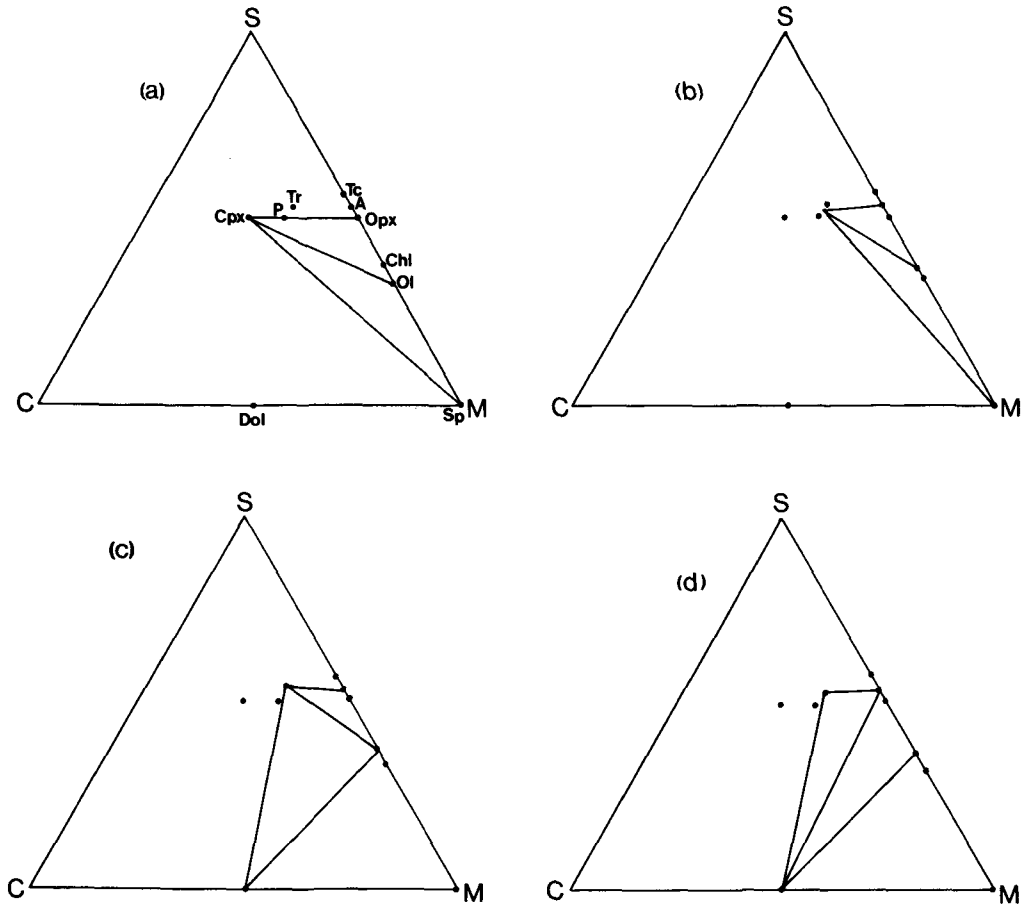


Fig. 2. Compatibility diagrams illustrating the mineral assemblages described in the text. (a) granulite facies, (b) transitional phase between granulite facies and (c), (c) Inverian assemblages, (d) Laxfordian shear zone assemblages. All samples contain an Fe-oxide phase. Abbreviations are: dol—dolomite, sp—spinel, ol—olivine, opx—orthopyroxene, A—anthophyllite or cummingtonite, chl—chlorite, Tc—talca, Tr—tremolite, P—pargasite, and cpx—clinopyroxene. S—SiO₂, C—CaO, M—MgO+FeO, all projections are from Al₂O₃+H₂O+CO₂.

TABLE I. Possible reactions to form Inverian and Laxfordian assemblages

4 parg + 11 opx + 10 ol + 2 sp + 32H ₂ O	= 4 trem + 8 chl + 2Na ₂ O	(1)
4 parg + 9 opx + 8 ol + 24H ₂ O	= 4 trem + 6 chl + 2Na ₂ O	(2)
parg + 2 ol	= trem + magt	(3)
4 cpx + 7 opx + 2 sp + 10H ₂ O	= 2 trem + 2 chl	(4)
12 opx + 3H ₂ O	= 3 anth + magt	(5)
opx + ol + sp + 4H ₂ O	= chl	(6)
6 trem + magt + 24CO ₂	= 3 anth + 12 dol + 24 SiO ₂ + 3H ₂ O	(7)
trem + magt + 2CO ₂	= anth + 2 cc	(8)
2 trem + 7CO ₂	= 3 dol + cc + anth	(9)

Abbreviations as follows: parg—pargasite, NaCa₂(Mg,Fe)₄Al₃Si₆O₂₂(OH)₂; opx—orthopyroxene, (Mg,Fe)₂Si₂O₆; cpx—clinopyroxene, Ca(Mg,Fe)Si₂O₆; ol—olivine, (Mg,Fe)SiO₄; sp—spinel, (Mg,Fe)Al₂O₄; magt—magnetite, Fe₃O₄; dol—dolomite, CaMg(CO₃)₂; cc—calcite, CaCO₃; trem—tremolite, Ca₂(Mg,Fe)₅Si₈O₂₂(OH)₂; anth—anthophyllite, (Mg,Fe)₇Si₈O₂₂(OH)₂; chl—chlorite, (Mg,Fe)₅Al₂Si₃O₁₀(OH)₈.

TABLE II. *Representative mineral analyses*

	W1 parg	W49 hb	D25 trem (a)	D25 trem (b)	D26 chl	W9 chl	W9 anth
SiO ₂	42.46	43.91	55.61	55.48	29.59	30.98	57.41
TiO ₂	1.53	0.40	0.06	0.09	0.05	0.05	0.03
Al ₂ O ₃	13.47	14.72	1.44	1.91	18.84	17.75	0.57
FeO	6.60	8.59	5.15	5.34	6.31	4.96	10.70
MnO	0.08	0.14	0.17	0.19	0.02	0.03	0.56
MgO	16.68	15.26	21.47	21.59	28.93	30.71	25.92
CaO	12.29	12.31	12.45	11.70	0.07	0.21	0.83
Na ₂ O	2.56	2.13	0.29	0.46	0.03	0.05	0.00
K ₂ O	0.64	0.34	0.02	0.04	0.03	0.00	0.00
Cr ₂ O ₃	0.32	0.02	0.05	0.16	0.81	0.25	0.01
NiO	n.d.	n.d.	n.d.	n.d.	0.23	0.21	0.11
Total	96.63	97.82	96.71	96.96	84.90	85.04	96.14
No. of oxygens	23	23	23	23	28	28	23
Si	6.114	6.313	7.799	7.754	5.826	6.029	8.006
Al ^{iv}	1.816	1.687	0.201	0.246	4.370	4.070	
Al ^{vi}	0.495	0.806	0.036	0.069			0.093
Ti	0.168	0.043	0.006	0.009	0.008	0.007	0.003
Cr	0.036	0.002	0.006	0.018	0.126	0.025	0.001
Fe	0.804	1.033	0.604	0.624	1.040	0.806	1.248
Mn	0.010	0.017	0.021	0.023	0.003	0.004	0.066
Mg	3.621	3.270	4.488	4.499	8.484	8.907	5.389
Ca	1.918	1.895	1.871	1.755	0.016	0.010	0.012
Na	0.724	0.595	0.078	0.124	0.010	0.005	0.000
K	0.120	0.063	0.004	0.008	0.006	0.000	0.000
Mg							
Mg + Fe	0.816	0.760	0.878	0.874	0.891	0.917	0.812

Abbreviations as in Table I. n.d.—element not determined.

W1, D25, D26, and W9 are from the Drumberg body (NC117333); W49 is a partially retrogressed granulite from Gorm Loch, north-east of Scourie (NC215445). In sample D25, trem (a) is a euhedral grain and trem (b) is a fine grain from a trem-chl intergrowth.

the University of Leicester. A range of natural and synthetic mineral and pure metal standards were used. Dead time and full ZAF corrections were applied using the computer program MAGIC IV (Colby, unpubl. ms). Representative mineral analyses are given in Table II.

The details of the granulite-facies mineralogy are given in Savage and Sills (1980). Coexisting pyroxene pairs indicate equilibration between 800 and 900 °C.

Amphibole. The range in calcic amphibole composition is given in fig. 3. The granulite-facies grains are pargasite with moderate TiO₂ contents (up to 1.6 wt. %). When retrogression begins, the grains exsolve opaque oxides with a corresponding drop in the TiO₂ and Cr₂O₃ content of the host grain (e.g. W49; Table II). Inverian tremolites have up to 2.0 wt. % Al₂O₃ and low alkalis. The Cr₂O₃ content is variable with euhedral grains having Cr₂O₃ below the detection limit of 0.05 wt. %, while

the fine grained tremolite has up to 0.8 wt. % Cr₂O₃. Anthophyllite and cummingtonite have less than 1.0 wt. % Al₂O₃ and CaO and are more iron-rich than coexisting tremolite. Tremolite and chlorite are both more magnesian than the granulite-facies minerals, which is a result of the increase in modal magnetite. The cummingtonite from shear zone assemblages has a lower Mg/(Mg+Fe) ratio than the Inverian tremolite and is associated with a slight reduction in modal Fe-oxide content, perhaps due to reactions such as 7 and 8 (Table I).

Chlorite. Chlorite is an Mg-rich clinocllore. The Al₂O₃ content increases from the first appearance of chlorite in Inverian assemblages to that found in Laxfordian shear zones, ranging from 3.5 to 4.5 atoms of Al per 28 oxygens. Chlorite has up to 1.0 wt. % Cr₂O₃ and 0.25 wt. % NiO. The Mg content of the minerals is such that $X_{Mg}^{chl} > X_{Mg}^{trem} > X_{Mg}^{anth}$.

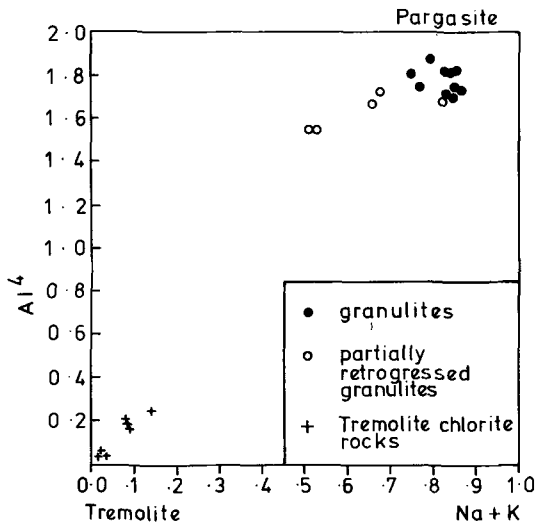


FIG. 3. Plot Al^{iv} against $Na+K$ for amphiboles from granulite-facies and retrogressed assemblages. Cations recalculated on the basis of 23O in water-free formula.

Carbonate. The dominant carbonate mineral is dolomite with a $Ca:Mg:Fe$ ratio of 50:45:5.

P-T conditions during retrogression

The pressure and temperature of retrogressive metamorphism can be assessed semi-quantitatively by reference to experimentally determined phase relationships in the pure $MgO-SiO_2-H_2O$ and $MgO-CaO-SiO_2-H_2O$ systems. It is difficult to make a precise estimate of the $P-T$ conditions because of the high variance of the Inverian and Laxfordian assemblages and the lack of accurate thermodynamic data, especially for cummingtonite.

The first mineral assemblage to form (trem + chl + anth) is entirely hydrous whilst in and near shear zones and dyke margins dolomite becomes significant in ultramafic rocks and calcite is locally abundant in mafic and intermediate rocks. This indicates that the fluid phase became more CO_2 -rich with time. The high variance of the assemblages suggests that the composition of the fluid was externally buffered, rather than controlled by the mineral assemblage. In this case it is likely that $P_{fluid} = P_{total}$, especially as retrogression occurred over a large area.

The increase in abundance of magnetite, the presence of hematite in shear zone assemblages and the increase in Fe^{3+} content of hornblendes from mafic and intermediate rocks (Sills, 1981) all indicate an increase in f_{O_2} associated with retrogression. An increase in f_{O_2} has also been reported from the

Scourie region (Beach and Fyfe, 1972; Rollinson, 1980).

The stability of anthophyllite has received a considerable amount of attention (Greenwood, 1963, 1971; Johannes, 1969; Day and Halbach, 1979), partly because of its limited stability. In the pure $MgO-SiO_2-H_2O$ system it is stable between approximately 650 and 810 °C (Day and Halbach, 1979), although the lower limit is very poorly constrained. It is stable down to 510 °C if CO_2 becomes the major proportion of the fluid phase (Johannes, 1969) and the presence of Fe is thought to lower the stability field to 600 °C at 7 kb (Evans and Trommsdorff, 1974). Mg-chlorite breaks down, in the pure Mg system, at about 800 °C at 6 kb (fig. 4; Fawcett and Yoder, 1966); 10–20% of the Fe end-member lowering this equilibrium by about 20 °C (Frost, 1975). Thus the initial trem + chl + anth assemblage indicates that retrogression began at temperatures between 600 and 800 °C. The lack of serpentinization, except as a late-stage phenomenon, suggests that temperatures remained above 480 °C (Johannes, 1969), although during Laxfordian shearing P_{CO_2} was high enough for serpentine to be unstable at any temperature.

In order to further constrain the $P-T-X_{H_2O}$ conditions of retrogression, several reaction boundaries have been calculated (fig. 4; reactions 1, 4, 7, and 8 from Table I). The thermodynamic data was taken from Powell (1978) and Helgeson *et al.* (1978). The values for anthophyllite were used for cummingtonite. The imprecision of the thermodynamic data leads to considerable errors in the calculated boundaries. The reactions are plotted for the natural mineral compositions, with K_D calculated assuming an ideal mixing on sites model for all minerals. This lowers the boundaries by about 30 °C from the end-member reactions. The fluid phase is assumed to be an ideal mixture of H_2O and CO_2 , such that $X_{H_2O} + X_{CO_2} = 1$.

Fig. 4a shows that trem + chl is stable up to about 775 °C with $P_{H_2O} = P_{TOT}$. Reduction of the activity of H_2O to 0.5 lowers the reactions by about 100 °C. Reactions 1 and 4 and the chlorite breakdown reaction are fairly insensitive to pressure. The tremolite-out reactions (7 and 8, fig. 4c) show that dol + anth is stable up to 800 °C when $X_{H_2O} < 0.8$; when X_{H_2O} is increased tremolite is stable to lower temperatures. There is no evidence for partial melting of the acid and intermediate gneisses during the Inverian, therefore the temperature must have been less than the granite minimum melting curve (650 °C at 6 kb, $P_{H_2O} = P_{TOT}$; Winkler, 1976). Temperatures of about 600 °C for the Inverian are also indicated by garnet-biotite pairs and feldspar thermometry (Sills, 1981) and by oxide thermometry (Rollinson, 1980).

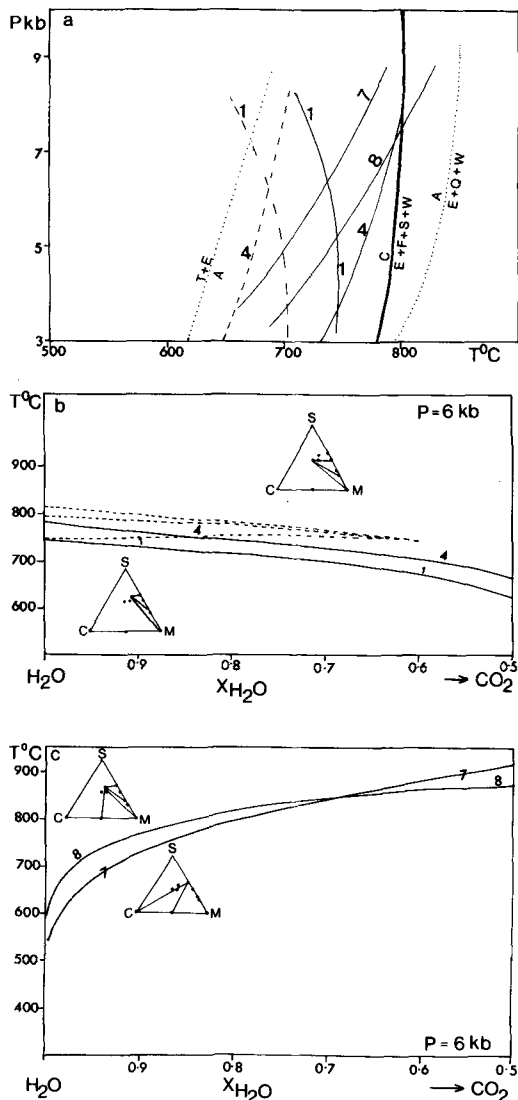


FIG. 4. (a) P - T plot for various reactions. Reactions labelled 1, 4, 7, and 8 are from Table I. For reactions 1 and 4 solid lines are for $P_{H_2O} = P_{TOT}$, dashed lines for $X_{H_2O} = 0.5$. Reactions 7 and 8 are calculated using $X_{H_2O} = 0.9$, $X_{CO_2} = 0.1$. The thick line is for the experimentally determined breakdown of magnesian chlorite (C) to enstatite (E) quartz (Q) and water (W) (Fawcett and Yoder, 1966). The dotted lines are for the stability field of anthophyllite (Day and Halbach, 1979) in the pure $MgO-SiO_2-H_2O$ system. (b) T - X_{CO_2} plot for reactions 1 and 4. Compatibility diagrams as for fig. 2. The dashed lines are for the reactions $Tc + E = A$, $Tc = A + E$ and $A = E + Q$ (abbreviations as above and fig. 2) at 7 kb after Greenwood (1963) and Johannes (1969). The location of the lower reaction is very uncertain. (c) T - X_{CO_2} plot for reactions 7 and 8. Compatibility diagrams as for fig. 2.

Fig. 4c indicates that $X_{H_2O} > 0.95$ for tremolite to be the stable Inverian phase rather than anth + dol at 600–650°C. To form the Laxfordian shear zone assemblage (cumm + dol + cc), X_{CO_2} was increased slightly.

In summary the possible P - T evolution of the ultramafic rocks is as follows:

(a) Metamorphism at granulite facies at a P - T of 10–14 kb and 800–900°C (Savage and Sills, 1980).

(b) Beginning of retrogression to form anth + trem + chl assemblages at temperatures between 600 and 800°C.

(c) Development of Inverian trem + chl assemblages with temperatures around 550–650°C and $X_{H_2O} > 0.9$.

(d) Shearing and recrystallization in Laxfordian shear zones with the breakdown of tremolite to form cumm + dol ± cc due to a slight increase in P_{CO_2} , possibly accompanied by a fall in temperature.

Throughout the retrogression the fluid phase was dominantly hydrous with the proportion of CO_2 increasing slightly in Laxfordian shear zones, although the proportion of CO_2 need not have exceeded 5% of the fluid phase. The pressure is very hard to constrain but was probably in the range 4–8 kb.

Changes in whole-rock composition during retrogression

It is interesting to consider whether any changes in bulk composition occurred during retrogression apart from the obvious addition of H_2O and CO_2 . From fig. 5 it can be seen that the trem + chl assemblages tend to have higher SiO_2 content than the granulite-facies samples. If the analyses were recalculated to 100%, the effect would be greater. No analyses are available for the cumm + dol + chl schists, but these must have significantly lower SiO_2 contents. In the $MgO-CaO-Al_2O_3-SiO_2$ system all the analyses fall within a volume bounded by anthophyllite, tremolite chlorite, and dolomite suggesting that no major composition change is required to develop that assemblage from the granulites. However, all retrogressed samples have very low Na + K (fig. 5) contents when compared with granulite-facies samples. In the granulites, Na + K is held in pargasite, whilst the Inverian amphibole, tremolite, has a very low alkali content. Na + K must therefore be lost from the ultramafic rocks during retrogression, but they need not have been totally removed as all other rock types seem to have gained Na during retrogression (Beach and Tarney, 1978).

Retrogression of all gneiss types occurred over a large part of the Assynt region indicating the

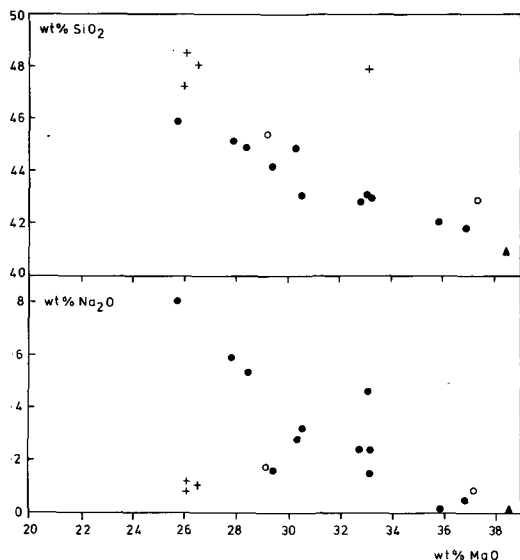


FIG. 5. Plot of SiO_2 and Na_2O against MgO for ultramafic rocks. Key as in fig. 2, with a triangle representing a serpentinite. Full analyses in Sills (1981).

presence of a pervasive fluid phase, which was dominantly hydrous. This enabled ions to be redistributed over large distances (Beach and Tarney, 1978) and caused an increase in the oxygen fugacity (Rollinson, 1980). The source of the fluids is uncertain, but the amounts required are quite large (1 km³ of water is needed to retrogress the approximately 50 km² of retrogressed rocks in Assynt assuming they are 1 km thick). The retrogression was intimately associated with the formation of north-west-trending structures which acted as channel-ways for the fluids.

In summary, the retrogression was caused by the influx of large volumes of dominantly hydrous fluids. The granulites were altered to assemblages dominated by tremolite and chlorite to be replaced by cummingtonite and dolomite assemblages in Laxfordian shear zones. The estimated temperatures suggest that this occurred in the middle crust.

Acknowledgements. The author would like to thank R. N. Wilson for help with the microprobe analyses and G. Hendry at the University of Birmingham for running the XRF analyses. This work was undertaken during the receipt of an NERC Research Studentship which is gratefully acknowledged and B. F. Windley is thanked for supervision.

REFERENCES

- Beach, A., and Fyfe, W. S. (1972) *Contrib. Mineral. Petrol.* **36**, 175.
 — and Tarney, J. (1978) *Precamb. Res.* **7**, 325.
 Bowes, D. R., Wright, A. E., and Park, R. G. (1964) *J. Geol. Soc. Lond.* **120**, 153.
 Chapman, H. J., and Moorbath, S. (1977) *Nature* **268**, 41.
 Day, H. W., and Halbach, H. (1979) *Am. Mineral.* **64**, 809.
 Evans, B. W., and Trommsdorff, V. (1974) *Am. J. Sci.* **274**, 274.
 Fawcett, J. J., and Yoder, H. S. (1966) *Am. Mineral.* **51**, 353.
 Frost, R. B. (1975) *J. Petrol.* **16**, 272.
 Greenwood, H. J. (1963). *Ibid.* **4**, 317.
 — (1971) *Am. J. Sci.* **270**, 151.
 Helgeson, H. C., Delaney, J. M., Nesbitt, H. W., and Bird, D. K. (1978) *Ibid.* **278-A**, 1.
 Johannes, W. (1969) *Ibid.* **267**, 1083.
 Powell, R. (1978) *Equilibrium thermodynamics in Petrology* (Harper and Row, London).
 Rollinson, H. R. (1980) *Mineral Mag.* **43**, 623.
 Savage, D., and Sills, J. D. (1980) *Contrib. Mineral. Petrol.* **74**, 153.
 Sheraton, J. W., Tarney, J., Wheatley, T. J., and Wright, A. E. (1973) In Park, R. G., and Tarney, J. (eds). *The Early Precambrian of Scotland and Related Rocks of Greenland*. (Keele Univ. Press), 31.
 Sills, J. D. (1981) unpubl. Ph.D. Thesis. Univ. of Leicester.
 Tarney, J. (1973) In Park, R. G., and Tarney, J. (eds). *The Early Precambrian of Scotland and Related Rocks of Greenland* (Keele Univ. Press), 105.
 Winkler, H. G. F. (1976) *Petrogenesis of metamorphic rocks*. 4th edition. (Springer-Verlag).

[Manuscript received 5 May 1981;
 revised 19 August 1981]