Graphite-bearing peraluminous dacites from the Erlend volcanic complex, Faeroe-Shetland Basin, North Atlantic

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Abstract

Strongly peraluminous, cordierite-bearing anatectic dacites from the offshore Tertiary Erlend volcanic centre, north of the Shetland Isles, are shown to contain graphite which is interpreted as being essentially a restite phase inherited from carbonaceous pelitic source rocks. The form and characteristics of the graphite are documented and graphite geothermometry applied to establish that the graphite records a minimum peak temperature of ~800°C, confirming that temperatures at which anatexis occurs were attained. The different morphological forms of graphite observed suggest the possibility that minor amounts of fluid-deposited graphite may also be present. The chemistry of the Erlend dacites is compared with that of other known examples of graphite-bearing peraluminous silicic igneous rocks and briefly with experimentally generated peraluminous liquid compositions. The Erlend source rocks were probably subjected to a higher degree of partial melting than has occurred in the petrogenesis of many other anatectic peraluminous silicic rocks.

KEYWORDS: graphite, peraluminous, dacite, anatectic, North Atlantic.

Introduction

A recent mineralogical and chemical study (Kanaris-Sotiriou *et al.*, 1993) of volcanic rocks recovered in three cores from well 209/3-1 (Fig. 1) drilled close to the centre of the Erlend volcanic complex, in the Faeroe-Shetland Basin, revealed a sequence of Tertiary MORB-type tholeiitic basalts lying above highly peraluminous cordierite-bearing dacites containing graphite. On the basis of the chemical, mineralogical and isotope data it was concluded that the dacites formed by low-pressure crystallisation of melts produced by anatexis of carbonaceous crustal material — possibly Cretaceous sediments forming the basin-fill. The Erlend centre is one of the three localities in the North Atlantic now known to have the same unusual bimodal Tertiary volcanic sequence of MORB-type basalts overlying peraluminous dacites, other examples having been described from the Rockall Trough (Morton *et al.*, 1988) and the Vøring Plateau (Parson *et al.*, 1989).

Although graphite occurs widely in many rock types, including plutonic igneous rocks, published accounts of the occurrence of graphite in *volcanic* rocks and minor intrusions where the origin and incorporation of the graphite can be related to the anatectic process are relatively rare, only three examples having been found among relatively recent literature. The major graphite occurrence at Seathwaite, Cumbria, although located within rocks of the Borrowdale Volcanic Group, appears to have been introduced, by replacement processes, into an intrusive diabase host via hydrothermal fluids from underlying carbonaceous mudstones (Strens, 1965; Weiss *et al.*, 1981).

Zeck (1970,1992) described an anatectic S-type dacite from Cerro del Hoyazo, south-eastern Spain, in which graphite crystals occur in the groundmass as a 'monocrystal' restite phase. These are found along with other restite components, including monocrystal inclusions of cordierite and almandine-rich garnet. Al-rich restite rock inclusions forming 10–15% of the dacite represent the (semi-) pelitic precursor from which the dacitic magma had formed by anatexis. The dacite also contains inclusions of gabbroic and basaltic lithologies which themselves contain restite inclusions similar to those found in the dacite matrix. Zeck (1970) offers two petrogenetic scenarios for the

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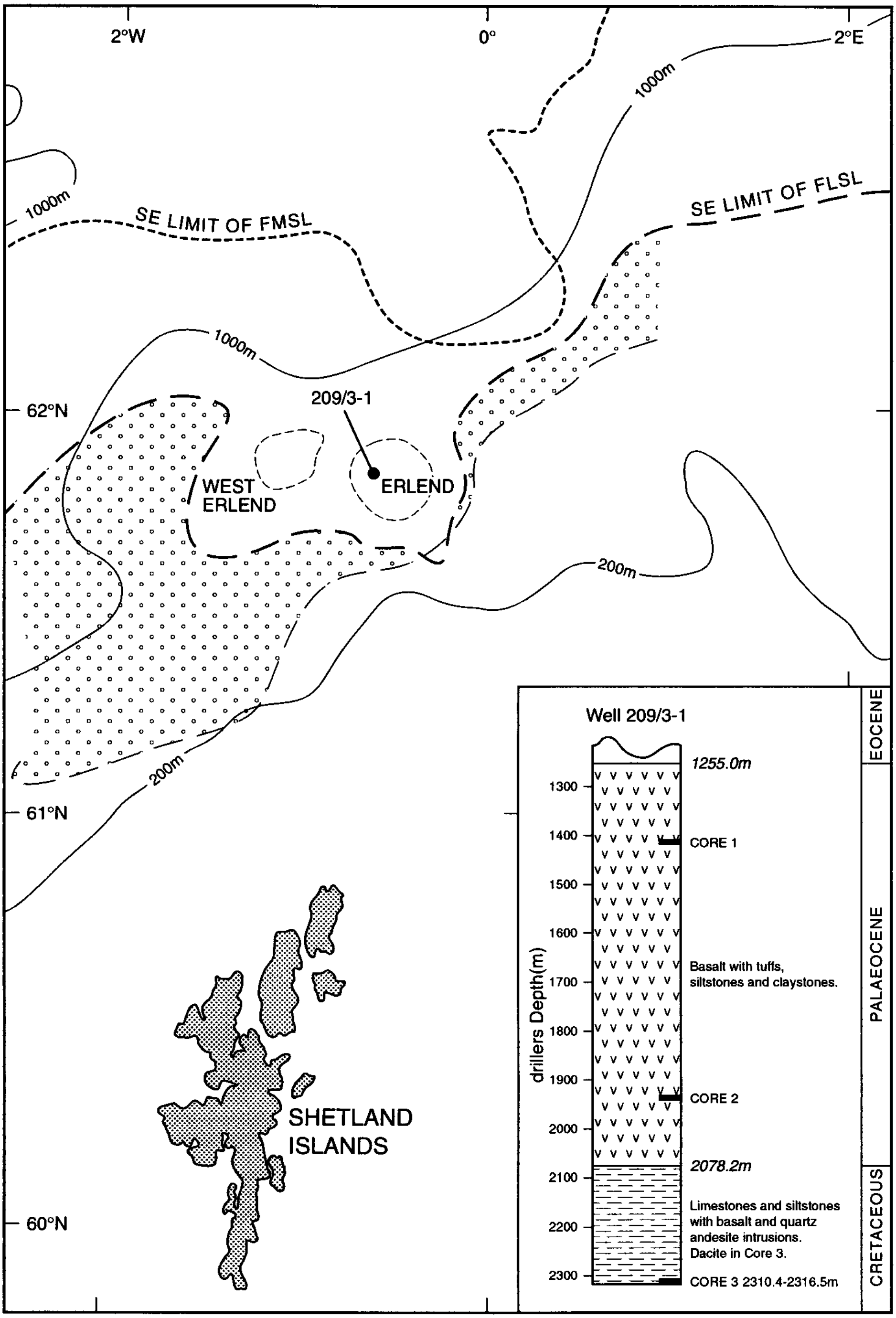

FIG. 1. Submarine geology of the Erlend and West Erlend volcanic centres after Gatliff *et al.* (1984) with boundaries of Faeroes Middle Series lavas (FMSL); Faeroes Lower Series Lavas (FLSL). Submarine contours are shown as solid lines. Stippled area, Faeroe-Shetland intrusive complex, SE limit after Ridd (1983). Inset — schematic of part of 209/3-1 well log after Kanaris-Sotiriou *et al.* (1993).

anatectic origin of the dacite, the first involving regional anatexis and the second invoking partial melting of a migmatite complex by intrusion of basic magma. The graphite crystals in the dacite are suggested to have been derived largely from graphite-bearing metamorphic rock inclusions.

Luque *et al.* (1987, 1993) documented graphite-bearing silicic dykes associated with marginal zones of ultramafic massifs in the Serranía de Ronda (Southern Spain). The dykes contain cordierite, quartz, garnet, biotite, ilmenite and graphite, the last occurring as randomly distributed, platy, xenomorphous aggregates with textural relations that indicated synchronous formation with the other minerals present. The graphite was thought to have been formed from CO_2- and CH_4-rich fluids originating during the anatexis of carbonaceous sedimentary rocks underlying the ultramafic rocks.

Pedersen (1981) gave an account of native iron-bearing andesitic-dacitic lavas from Disko, West Greenland, that equilibrated under reducing f_{O_2} conditions controlled by the presence of carbon incorporated into the magma from the source rocks. Graphite is present in small quantities in some of the rocks, where it mostly occurs enclosed in oxide phenocrysts. The volcanic rocks also contain complex Fe-Ti oxides-sulphides, including armalcolite, similar to lunar type 1 armalcolite. In the latter paper, the volcanic rocks described were identified as "the most silicic volcanic rocks with native iron yet known from Earth". Pedersen suggested a petrogenesis for the Disko silicic rocks that involves extensive reaction between tholeiitic basalt magma and carbonaceous sediments at pressures of about 1–1.5 kbar, the contaminated magmas generated eventually ascending through the lava pile and being erupted as lavas and tephra.

This paper gives the evidence for the presence and characterisation of the graphite in the Erlend dacitic rocks from Core 3 (Fig. 1) of well 209/301. The petrogenetic significance of the occurrence of graphite in these rocks is considered, together with a comparison with the other examples of graphite-bearing anatectic dacitic rocks mentioned.

Petrography and graphite paragenesis

The Erlend dacites are glasses that are variably devitrified and contain up to 10% cordierite or pinitic mica pseudomorphs after cordierite. X-ray diffraction (XRD) analysis of the dacite whole-rock indicated the dominant silicate minerals to be plagioclase, quartz, chlorite and muscovite/illite. During the preparation of smear specimens for XRD investigation, it was noticed that one of the dacite samples in particular (sample 3-3, which contains 0.61% C) produced a graphite-like surface film when the sample was ground with water. Separation of the material forming the suspended film, and subsequent XRD analysis identified the material positively as graphite (Fig. 2). All the dacite samples, in fact, contain more than 0.1% carbon in a non-carbonate form and it therefore seems likely that most of this carbon is present in the form of graphite.

A variety of other minor opaque phases apart from graphite are present. Electron microprobe analysis confirmed that these include pyrite, ilmenite and possibly pyrrhotite, but no metallic iron has been detected.

Optical examination of a thin section of sample 3-3 revealed the presence of rounded- to irregularly-shaped aggregates, up to five millimetres or so in diameter, consisting mainly of flakes (up to about 0.1 mm in length and 0.01 mm wide) of opaque material with a strongly cleaved habit that is compatible with that of graphite (Fig. 3*a*). The graphite appears to be confined to these aggregates. Some graphite crystals have structures (Fig. 3*b*) that probably represent graphite deformation effects. Small spheres of graphite with an internal radial structure have been observed in some of the inclusions (Fig. 3*b*). The measured reflectance of the graphite flakes is variable from ~4 up to ~7% $R_{random/oil}$, which is low compared with the maximum reflectance of graphite of 17.8% R_{max} (Stach *et al.*, 1982, p. 46), but this may, in part, be due to the narrowness of the graphite flakes compared with the reflectance light spot. Also, the reflectance of graphite is known to vary with form, the transition from semigraphite to graphite being characterised by reflectances of >5% R_{max}.

None of the modes of occurrence of graphite cited earlier, in which the graphite is dispersed throughout the rock, resembles that found in the Erlend dacites, in which the graphite is not distributed randomly throughout the groundmass, but appears to be restricted to isolated, more or less rounded aggregates of numerous graphite crystals. Although graphite can be fluid-deposited (see Luque *et al.*, 1995), it is difficult to envisage inclusions of the form seen in the Erlend rocks being introduced in this way and hence they would appear to be most realistically interpreted as small residual enclaves (restite) of metamorphosed carbonaceous sedimentary rock that presumably was the source of the anatectic dacitic melt.

Although spherulitic forms of graphite have been described in 'titanomagnetite differentiates' in magmatic rocks illustrated by Ramdohr (1980; p. 391; Fig. 325), this morphology has more recently been interpreted as representing graphite that has been fluid-deposited. Duke and Rumble (1985) describe spherulitic graphite in granites from New Hampshire as forming from late-stage C-O-H fluids which were also responsible for silicate alteration. These authors also observed that such spherules may nucleate on pre-

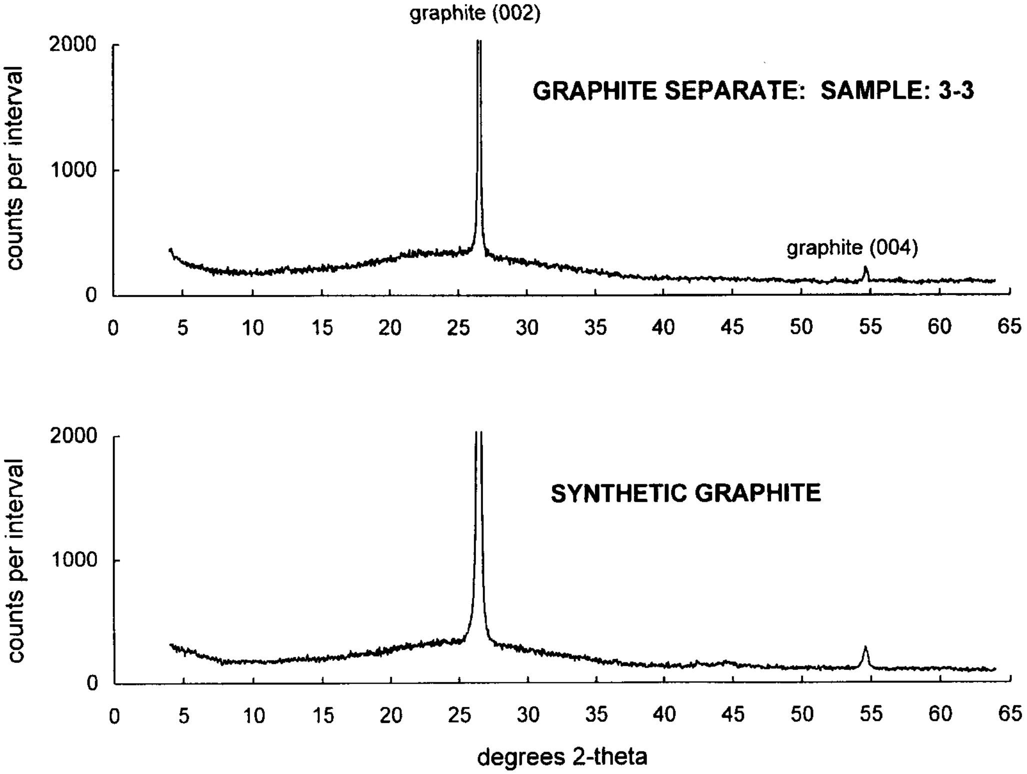

FIG. 2. X-ray diffraction patterns of (above) graphite separated from dacite sample 3-3 (see Table 1) and (below) synthetic graphite. Both patterns were from samples deposited on glass slides and obtained using Cu-*K*α radiation.

existing flake graphite. Fluid inclusion evidence for the influence of CO_2-rich fluids at both magmatic and post-magmatic stages of the development of graphite-bearing peraluminous granites in Anarctica was given by Frezzotti *et al.* (1994). They observed that, although graphite occurred in three different forms in the rocks studied, it was always present as a secondary phase, although it did not exhibit a spherulitic form.

The presence of spherules of graphite amongst aggregates of flake graphite in the Erlend rocks may, therefore, indicate that there could be more than one generation of graphite present, with the spherulitic graphite being deposited from a C-bearing aqueous fluid onto flake graphite which was of earlier metamorphic (restitic) origin. The presence of such a fluid would also be compatible with the the late-stage devitrification of the matrix glass and hydrous alteration of the cordierite phenocrysts.

Graphite geothermometry

Luque *et al.* (1993) studied graphites occurring in low- and high-temperature regimes (black shales and acid dykes) in Spain, applying the experimentally based graphite geothermometer of Shengelia *et al.* (1979) developed for progressively graphitized carbonaceous material (Fig. 4). They established that for rocks subjected to low-grade metamorphic conditions the graphite c_0 parameter (2 × the $d_{(002)}$ spacing) is affected by factors other than temperature. The most important of these factors appears to be shear stress, with variations in lithology and the nature of the original organic debris being of minor significance. At temperatures above 500°C, however, where fully ordered graphite (graphite giving an XRD pattern corresponding to the ideal graphite structure) first appears, a close correlation exists between temperatures obtained from the structural ordering parameters of graphite and those obtained from mineral exchange geothermometers. Luque *et al.* (1993 and *pers.com.*) conclude that graphite geothermometry (in which the c_0 parameter has been *directly* related to temperature) is useful in determining peak temperatures, both in the case of fluid-deposited graphite and of metamorphosed carbonaceous material, and that it is independent of

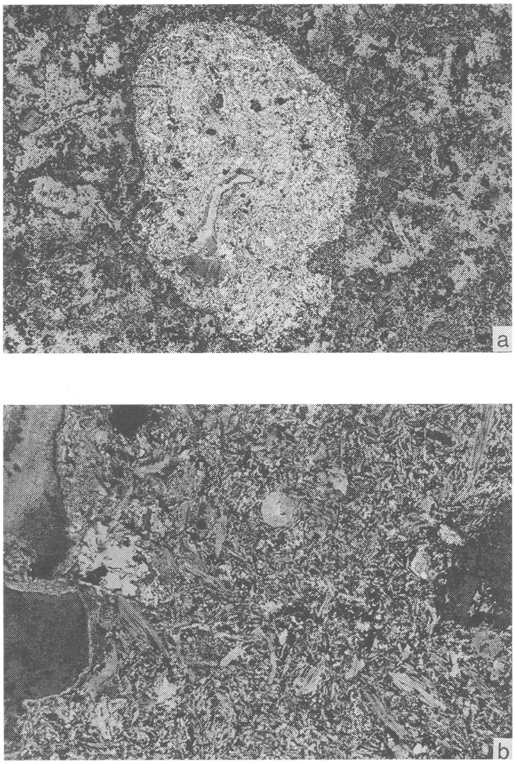

Fig. 3. Photomicrographs of a graphite inclusion (high reflectance) in dacite sample 3-3, taken in reflected light. In (*a*) the field width is 2.5 mm; in (*b*) the field width is 0.75 mm and is an enlarged view of an area in the lower part of the graphite inclusion shown in (*a*).

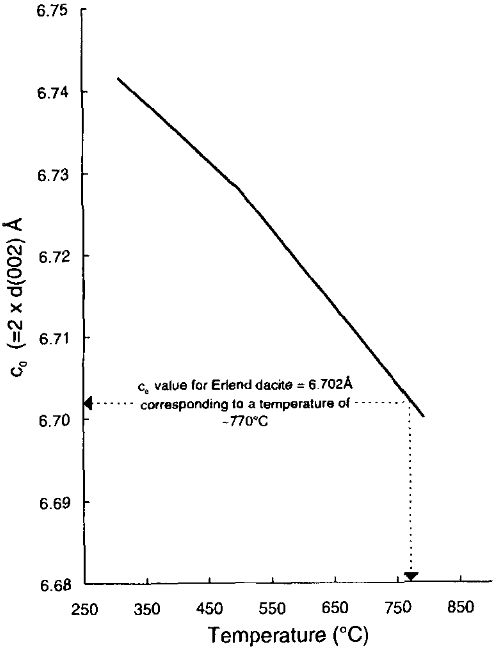

Fig. 4. Estimated temperature recorded by the graphite in the Erlend dacite, using the Shengelia *et al.* (1979) geothermometer calibration.

retrograde metamorphic effects. Differences between temperature estimations above 500°C by graphite geothermometry and exchange geothermometry were given as 10–40°C by Luque *et al.* (1993), who place the upper temperature limit of the graphite geothermometer at about 800°C. No data are available for graphites formed above this temperature. Luque *et al.* (1987, 1993) concluded that the graphite in the acid dykes they studied record peak temperatures (before hydrothermal alteration) of 825 ± 15°C. No significant difference between peak temperatures determined for unaltered dyke lithologies and those affected by hydrothermal alteration was detected. Plotting the c_0 parameter for graphite from the Erlend dacites (6.702 Å) on Fig. 4 indicates that a peak temperature of ~770°C (± say 40°C) was attained by these rocks.

Wada *et al.* (1994), by contrast, compare the graphitization of carbonaceous matter in carbonate and pelitic rocks affected by (a) regional metamorphism (Ryoke metamorphic terrain) and (b) contact metamorphism in the Kaizukiyama granite contact aureole of the Kasuga area, both in Central Japan. Applying XRD, DTA-TG analysis and chemical and stable isotope analyses, they demonstrated a relationship between the basal spacing (d(002)) and the crystallite size (Lc(002)) of the carbonaceous matter, and related metamorphic temperature independently derived from calcite–dolomite and carbon isotope geothermometers to relative degrees of graphitization (DG) calculated from XRD pattern parameters. The Kasuga pelites and limestones show inexplicably different linear relationships of DG to metamorphic temperature, although there is some uncertainty in the exact location of the Kasuga pelites more distant from the granite contact (Wada, *pers. com.*). Because the DG parameter is influenced by instrumental differences, such relationships cannot be reliably transferred to other geological examples, unlike the Shengelia *et al.* (1979) approach which is applied here.

The similar peak temperatures (~800°C) indicated for both the Erlend dacites and the acid dykes described by Luque *et al.* (1987) should be regarded as minimum temperatures, since they are at the upper limit of the graphite geothermometer. Partial melting

TABLE 1. Compositions of graphite-bearing anatectic dacites

	Erlend: Well 209/3-1[1]				Cerro del Hoyazo lava[2]	Serranía de Ronda dykes[3]		Disko lava[4]	*Macasuni ash-flow tuff*[5]
Sample	3-1	3-2	3-3	3-4		U	T		
wt.%									
SiO_2	64.14	62.75	65.01	65.87	63.67	58.26	52.05	66.20	73.1
TiO_2	0.84	0.80	0.85	0.84	0.68	0.80	0.24	1.25	0.14
Al_2O_3	18.08	18.50	16.85	16.94	17.12	19.14	17.56	13.59	14.9
Fe_2O_3	1.67	2.13	1.82	1.31	0.67				
FeO	3.28	3.32	3.34	3.60	4.18				
Fe_2O_3[t]	(5. 32)	(5.82)	(5.53)	(5.31)	(5.25)	11.57	10.85	7.08	1.37
MnO	0.08	0.09	0.13	0.11	0.09			0.10	0.03
MgO	1.99	2.08	1.43	1.62	1.84	2.95	3.31	1.29	0.20
CaO	0.99	1.10	2.01	1.91	2.47	1.20	4.43	3.62	0.60
Na_2O	0.57	0.54	2.02	1.56	2.21	0.18	0.98	2.67	3.23
K_2O	2.82	3.09	2.63	2.17	3.56	0.42	0.51	2.34	4.92
P_2O_5	0.07	0.12	0.13	0.12	0.21			0.31	0.42
S	0.02	0.10	0.05	0.03				0.28	
CO_2	0.05	0.12	0.04	0.05					
H_2O^+	4.56	4.84	2.89	3.16	2.27			1.09	
C	0.11	0.13	0.61	0.21	0.53			0.27	
L.O.I.						4.96	9.67		1.60
Total	99.53	99.71	99.90	99.50	99.50	99.44	99.60	100.09	100.51
					Selected CIPW norm constituents[6]				
%									
qz	43.35	40.79	35.15	40.52	27.77	43.98	25.23	30.50	34.61
c	12.53	12.79	7.36	8.91	5.64	16.21	7.34	0.80	4.18
or	16.71	18.31	15.58	12.86	21.04	2.48	3.01	13.85	29.08
ab	4.82	4.57	17.09	13.20	18.70	1.52	8.29	22.59	27.33
an	4.27	4.04	9.09	8.53	10.88	5.95	21.98	16.00	0.23

[1] Data for Erlend graphite-bearing dacites [core 3, well 209/31 (Fig.1)] from Kanaris-Sotiriou *et al*, 1993.

[2] Graphite-bearing dacite lava from Cerro del Hoyazo, southeastern Spain, (Zeck, H.P. (1970): Table 5, analysis Z 73 Ho).

[3] Graphite-bearing dacitic dykes from Serranía de Ronda, southern Spain (Luque, F.J., 1990).
U = unaltered; T = hydrothermally altered (mean of 10 samples in both cases).

[4] Graphite-bearing dacite from Disko, West Greenland (Pedersen, A.K. (1981): Table 1, analysis 4).

[5] Macasuni peraluminous ash-flow tuff (Pichavant *et al.* (1988*b*): Table 1, analysis MH4). [not graphite-bearing].

[t] total Fe expressed as Fe_2O_3; L.O.I. = loss on ignition.

[6] normative constituents calculated after normalising Fe_2O_3/FeO to 0.40 following Middlemost's (1989) recommendation for dacite norm calculation. A ratio of 0.50 was used for the Macasuni ash-flow tuff.

of a pelitic or semipelitic protolith would occur at this temperature and this is consistent with the proposed anatectic nature of the Erlend dacites (Kanaris-Sotiriou *et al.*, 1993).

Chemistry of the dacites

Major and trace-element data on the dacites were published by Kanaris-Sotiriou *et al.* (1993) who demonstrated the distinctive chemistry of the North Atlantic (Vøring Plateau, Rockall Trough and Erlend) dacites when compared with orogenic dacites. The dacites have average isotope ratios of $^{87}Sr/^{86}Sr_{60Ma} = \sim 0.71380$; $\varepsilon_{Nd} = -8.3$; $^{206}Pb/^{204}Pb = 18.538$; $^{207}Pb/^{204}Pb = 15.568$; $^{208}Pb/^{204}Pb = 38.573$, indicating that upper crustal material was involved in the origin of these rocks. The major-element data are reproduced here (Table 1) together with normative quartz and corundum, and corresponding data for the three other examples of graphite-bearing anatectic dacites cited in the literature and data for the peraluminous Macasuni volcanics (Pichavant *et al.*, 1988*b*) which are not reported to contain graphite (Pichavant *et al.*, 1988*a*). Chemical analysis of the

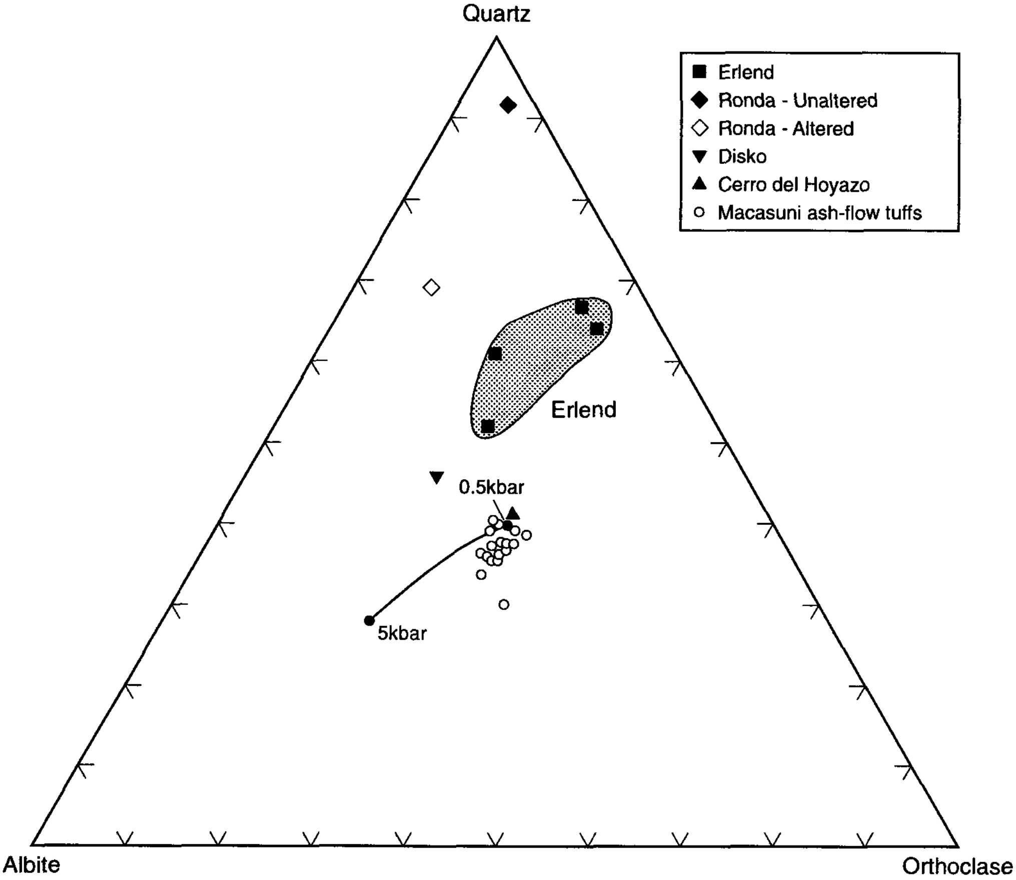

FIG. 5. Q-Or-Ab diagram for graphite-bearing dacites and the silicic Macasuni ash-flow tuffs (Pichavant *et al.*, 1988*b*). ● = H_2O saturated minima and eutectics at 0.5 kbar and 5 kbar (Luth, 1976).

Erlend dacites revealed a significant carbon content in four samples (0.11%; 0.13%; 0.61% and 0.21% — see Table 1). Although this carbon could be present as elemental carbon (including graphite), carbide or organically-bound carbon, the preceding part of this account demonstrates that the carbon is present in the form of graphite.

Comparison of the Erlend dacite compositions with melting point minima in the Qz-Or-Ab system and with other peraluminous rock compositions (Fig. 5) shows these rocks to be relatively enriched in quartz (or depleted in Ab and Or) compared with the minimum points, whilst the Ronda graphitic dacites contain even higher proportions of normative quartz. The Cerro del Hoyazo dacite lava composition, however, plots close to the low-pressure minimum (near to the Macasuni ash-flow tuffs; Pichavant *et al.* (1988*b*).

The Erlend dacites appear to be more peraluminous (average normative corundum = 10.40) and less silicic (average SiO_2 = 64.43%) than some other felsic volcanic rocks attributed to crustal melting (e.g. Pichavant *et al.*, 1988*b*) and to most S-type granitoids, but have a comparable major element chemistry to the two Spanish examples of graphite-bearing anatectic dacites which have significant normative corundum (in excess of 5%). The Erlend dacite samples, however, show some signs of alteration and mobility of some components may be anticipated.

It is argued, however, by Patiño Douce (1992) that chemical parameters such as the normative corundum content or Aluminium Saturation Index (ASI = $Al_2O_3/[CaO+Na_2O+K_2O]$) do not adequately reflect the activity of aluminium in silicic melts and that the approach used by Miller (1985) (a ternary plot of A = $Al_2O_3 - [CaO+Na_2O+K_2O]$; F = FeO; M = MgO) in establishing degrees of peraluminosity is preferable in this respect. Plotting the data for the Erlend dacites on the diagram suggested by Miller (1985) (Fig. 6) indicates these rocks to be strongly peraluminous (Ps) but possibly less so that the Macasuni ash-flow tuffs which, however, are more felsic lithologies (lower MgO and FeO). Therefore, whereas the normative corundum values of the Erlend rock suggest that these rocks are more peraluminous than the Macasuni lithologies, other approaches, whilst confirming the strongly peraluminous nature of both, suggest the Erlend rocks to be the less peraluminous.

Compared with other naturally occurring peraluminous compositions and with experimental peraluminous compositions (Green, 1976; Clemens and Wall, 1981; Vielzeuf and Holloway, 1988 (all relating to fluid-absent melting); Benard *et al.*, 1985 (H_2O-saturated melting)) when plotted on A'KF and ACF diagrams (see Pichavant *et al.*, 1988*b*), the Erlend dacites appear to represent distinctly different compositions (K-depleted and enriched in $MgO+FeO^t$), both with respect to the experimentally generated peraluminous liquids and to other natural peraluminous lithologies (e.g. Pichavant *et al.*, 1988*b*), but the Ronda dykes are even more distinctive, showing extreme depletion in K. (It is noted that there is no difference between the Ronda altered and unaltered samples in this respect).

Discussion

Whilst the chemistry of the Erlend dacites is closely similar to that of the garnet-bearing Cerro-del-Hoyazo dacitic lava (Table 1), the Erlend rocks do not contain garnet and therefore, on this basis, may not have reached the grade of the latter. However, compositional controls (low Fe/Mg ratio) may have

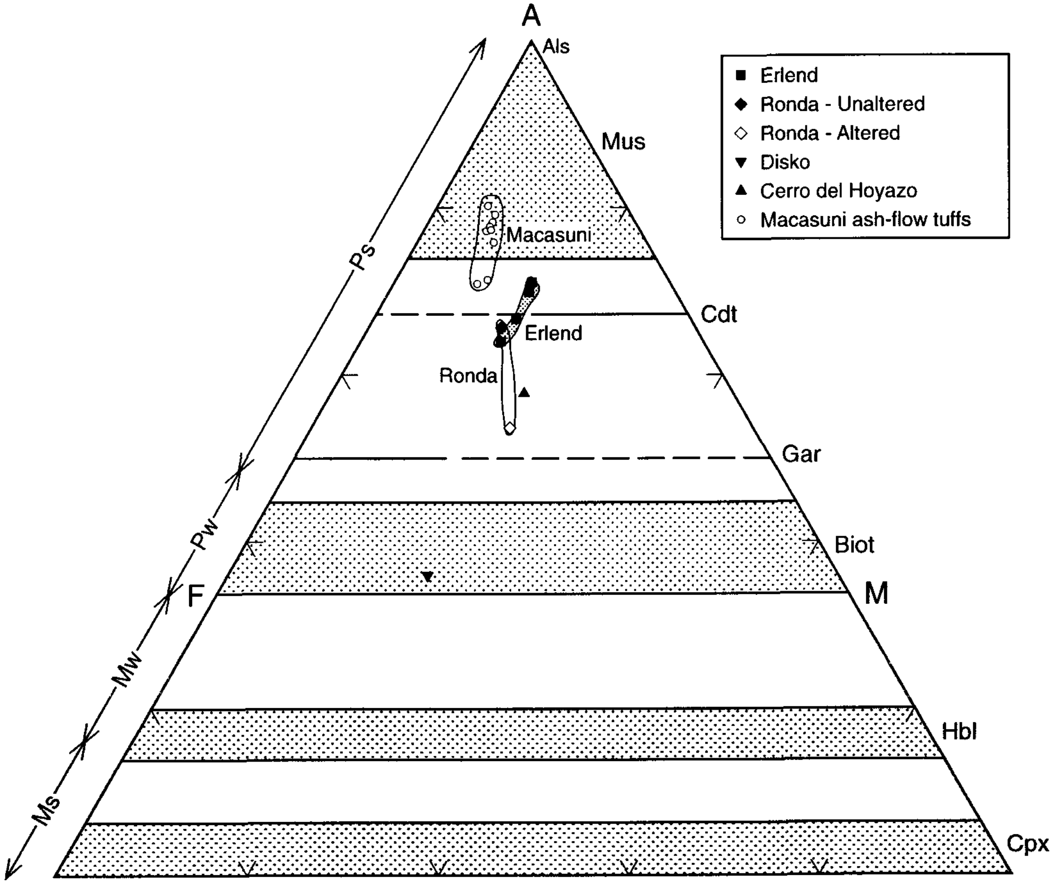

FIG. 6. AFM diagram after Miller, 1985. [A = Al_2O_3–[$CaO+Na_2O+K_2O$]; F = FeO; M = MgO]. Ps (strongly peraluminous) rocks contain minerals more aluminous than biotite. Pw (weakly peraluminous) are those containing biotite.

operated to prevent garnet formation in the case of Erlend rocks. The Serranía de Ronda dykes contain much more graphite (up to 50% — Luque *et al.*, 1993) than the Erlend dacites, have a correspondingly lower SiO_2 content and are also garnet-bearing, although they *may* have attained only marginally higher temperatures on the basis of the graphite geothermometer. The Disko dacites, by contrast, are only mildly peraluminous and have little or no normative corundum, although the suggested mechanisms for the origin of the Disko dacites are strikingly similar to those proposed for the dacites of the Erlend volcanic centre. The Disko lava also differs chemically in other respects (e.g. higher Ti and Ca contents and lower V and Cr).

The strongly peraluminous nature of both the Serranía de Ronda dykes and the Erlend dacites could reflect the influence of significant amounts of Al-rich restite inclusions on the bulk rock analysis of these lithologies. The compositions of the anatectic *melt* component of these examples and the Cerro del Hoyazo dacite *may* therefore be closer than the bulk analyses would indicate. The petrography of the Erlend dacites, however, does not show obvious inclusions of entrained aluminous restite material. This and their distinctive chemistry compared with examples of experimental peraluminous produced both by fluid-absent (dehydration) and fluid-saturated melting, suggests the possibility of non-minimum melting systematics and the further speculation that the strongly peraluminous nature of the rock may be due to exceptional degrees of partial melting of a carbonaceous pelitic protolith. The possibility, indicated by the presence of graphite in the rock, that the magma from which the Erlend dacites crystallised may have incorporated fluids with significant CO_2 or other C-O-H components that evolved in composition with cooling imposes further variables, the influence of which have yet to be fully clarified (Patiño Douce, 1992).

It is interesting that the most highly peraluminous examples of silicic magmatic rocks appear to occur where partial melting of pelite by basic magma is implicated. This suggests that the heat flux and/or temperature to which the source rocks were subjected as a result of the proximal emplacement of large volumes of basic magma is likely to be significantly greater than those involved in situations where S-type magmatic lithologies result from anatexis related to regional metamorphism.

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