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RESEARCH ARTICLE

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Mixed metamorphic and fluid graphite deposition in Palaeoproterozoic supracrustal rocks of the Lewisian Complex, NW Scotland

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Abstract

Graphite deposits may form alternatively by metamorphism of sedimentary rocks and from fluids. Both types occur in supracrustal successions within the Lewisian Complex of Northwest Scotland, and similarly in Palaeoproterozoic supracrustal rocks across the North Atlantic region in Canada, Greenland and Scandinavia. Carbon isotope compositions show that the graphite in Scotland had a mixed origin from metamorphism of sedimentary organic matter (schists) and the decarbonation of limestones (marbles). Raman spectroscopy shows that most of the graphite in Scotland exhibits some structural disorder, unlike the complete order in graphite vein ore deposits across the region. Exceptionally, where graphite was precipitated from fluid, in albitized rock in Tiree and Scardroy, it is fully ordered. While organic matter may survive granulite facies metamorphism without being transformed to fully ordered graphite, it can yield commercially more valuable ordered graphite when mobilized in a fluid.

KEYWORDS

carbon, carbon isotopes, graphite, Lewisian, marble, Palaeoproterozoic, Raman spectroscopy, Scotland

1 | INTRODUCTION

Graphite is a critical commodity because of the very high potential of graphite and graphene in future technologies, including its use in electric vehicles (Gautneb et al., 2019; Helmers, 2015; Wang et al., 2018). Exceptional demand has driven a revolution in graphite exploration, and the need to understand controls on graphite properties (Jara et al., 2019; Scogings, 2015). Most graphite resources occur in Precambrian rocks, reflecting the high incidence of black shales within the Precambrian (Condie et al., 2001) and the metamorphism of organic matter to graphite in older rocks.

Prospective graphite deposits have been explored in several parts of the North Atlantic region, including Labrador, Canada (Saglek Bay), Greenland (Amitsoq, Akuliaruseq), Norway (Skaland) and Sweden (Woxna). Each of these deposits was deposited during the period 1.8–2.1 Ga (Bergman, 2018; Meyer & Dean, 1988; Palosaari et al., 2016; Thrane & Kalvig, 2019). In the UK, the Lewisian of north-west Scotland includes several supracrustal successions (Figure 1), most of which contain graphite. Three of the supracrustal

successions have been dated at 1.8–2.1 Ga, like commercial deposits elsewhere. However, the graphite occurrences in the Lewisian are very poorly documented. We characterize graphite from six successions, using carbon isotope composition, Raman spectroscopy and microscopy, and investigate:

1. Is all the graphite derived from organic matter in shales, or is some derived from reduction of carbonate carbon in marbles in the supracrustal successions?
2. Is there evidence for mobilization of carbon from beds of carbon-rich sediment, i.e. graphite was deposited from a fluid phase rather than simply metamorphism of kerogen?
3. Is the carbon all fully ordered graphite, as required in graphite of commercial quality (e.g. Palosaari et al., 2020), or is some incompletely ordered?

Significance

Graphite is an underestimated component of supracrustal rocks in the extensively studied Lewisian Complex, and is shown to have two distinct origins.

2 | GEOLOGICAL SETTING

The Lewisian Complex of Northwestern Scotland consists predominantly of tonalitic gneisses of Archean age (Friend & Kinny, 1995), derived from an igneous protolith. However, there are several outliers of supracrustal rocks within the Lewisian, which represent

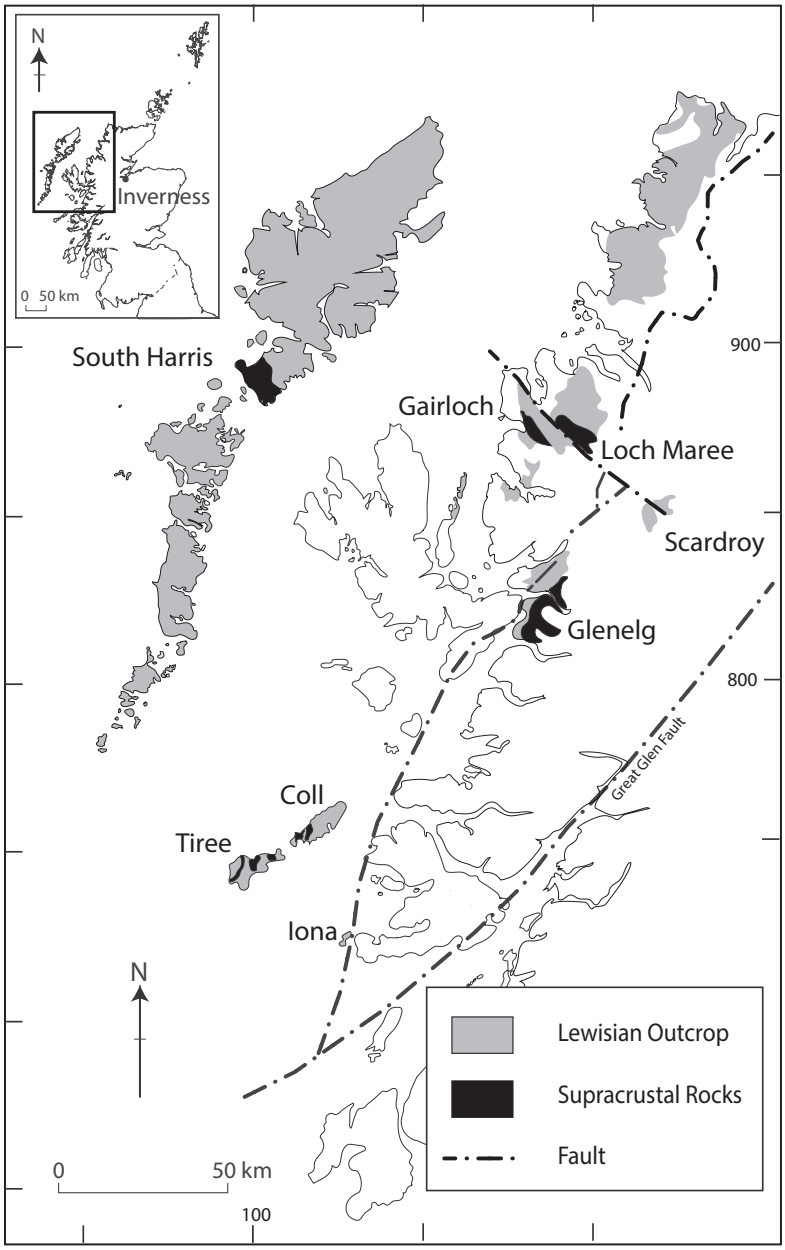

FIGURE 1 Map of North West Scotland, showing distribution of Lewisian supracrustal inliers. Graphite occurs in all named inliers [Colour figure can be viewed at wileyonlinelibrary.com]

amphibolite or granulite facies metasediments. The larger supracrustal outliers include carbonaceous (graphitic) pelites and schists, limestones represented as marbles, sulphide-rich horizons evident as rust zones, and non-sulphide iron formations. The combination of graphitic sediments, marbles and rust zones occur in each of six outliers: Gairloch-Loch Maree, South Harris, Tiree, Iona, Glenelg-Loch Duich and Scardroy (Figure 1). The similarities have been inferred as evidence that all are of similar age (Cartwright, 1992; Coats et al., 1997; Rock, 1987; Tilley, 1936). The outliers were formerly assumed to be in the main Lewisian succession, i.e. Archean (Cartwright, 1992; Whitehouse & Russell, 1997). Three of the six outliers now have age constraints in the mid-Palaeoproterozoic. The Loch Maree Group at Gairloch is dated 1.9–2.0 Ga, based on Nd crustal ages (O'Nions et al., 1983), minimum ages of detrital zircons (Kerr et al., 2016; Whitehouse et al., 1997) and a 1.90 Ga intrusive gneiss (Park et al., 2001). The metasediments in South Harris are dated 1.8–1.9 Ga, by detrital zircons (Whitehouse & Bridgwater, 2001) and associated ~1.9 Ga arc rocks (Mason et al., 2004). Eclogites at Glenelg-Loch Duich, whose protoliths were possibly synchronous with metasediments, yield Hf_{TDM} ages around 2.0 Ga (Brewer et al., 2003; Storey, 2008).

Graphite occurs in the supracrustal rocks in two distinct forms. Graphitic pelites and schists represent sedimentary rocks in which carbon was deposited as black shales. Graphite also occurs as laminae and nodules within marbles (Figure 2), deposited as limestones. In limestones, primary reduced organic matter was less likely, and the graphite may instead represent alteration of the limestone, or post-depositional introduction of carbon from shales elsewhere in the sequence. The graphite shows no relationship with major faults or other structures, but at two localities is associated with albitite veining.

The graphite in the six successions occurs as:

South Harris: Beds of graphitic schist, coloured silvery due to large flake size (< 2mm; Fettes et al., 1992).

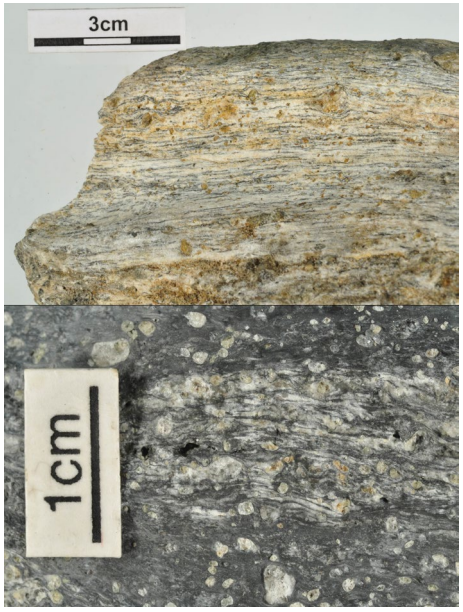

FIGURE 2 Graphite-bearing marble, Gott, Tiree. Laminate structure defined by graphitic streaks, moulded around phenocrysts [Colour figure can be viewed at wileyonlinelibrary.com]

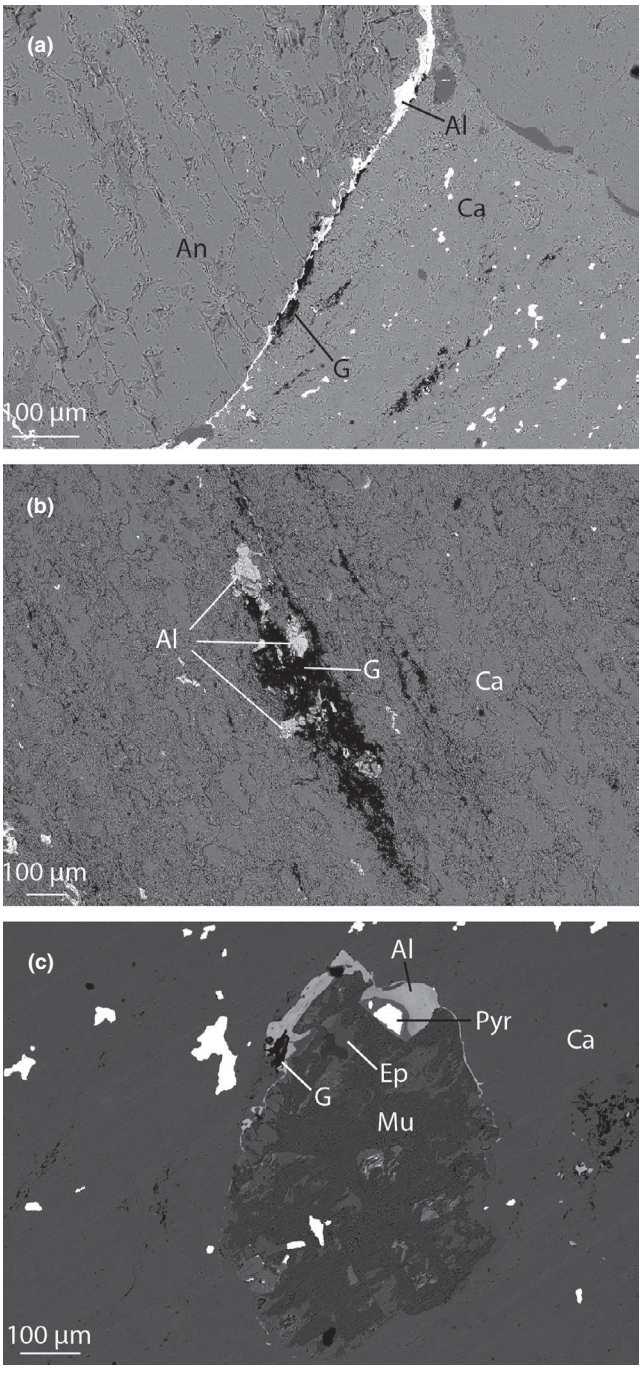

FIGURE 3 Backscattered electron micrographs of graphite in marble, Gott, Tiree. (a) Coating of graphite (G), and allanite (Al) around anorthite (An) phenocryst in marble (Ca); (b) Laminate streak of graphite (G) with intermixed crystals of allanite (Al) in marble (Ca); (c) coating of allanite (Al) and graphite (G) around phenocryst of muscovite (Mu) with epidote (Ep) and pyrite (Pyr) in marble (Ca) [Colour figure can be viewed at wileyonlinelibrary.com]

Gairloch-Loch Maree: Beds of graphitic schist, coloured grey to black (Park et al., 2001).

Glenelg-Loch Duich: Beds of graphitic schist, coloured silvery where well crystalline (flake size <5 mm), including adjacent to marble beds, to black where microcrystalline (Storey, 2008).

Scardroy: At least one bed of graphitic schist, and sub-millimetre nodules of graphite in marble (Rock, 1987; Sutton & Watson, 1951).

Tiree: Beds of graphitic schist, coloured black (section at Vaul) and marble containing intermittent laminae of graphite (Gott; Westbrook, 1972).

Iona: Beds of graphitic schist (flake size <2 mm) and marble containing laminae and patches of graphite (Bailey et al., 1925).

3 | METHODS

Scanning electron microscopy (SEM) was conducted in the Aberdeen Centre for Electron Microscopy, Analysis and Characterisation (ACEMAC) facility at the University of Aberdeen using a Carl Zeiss GeminiSEM 300 VP Field Emission instrument equipped with an Oxford Instruments NanoAnalysis Xmax80 Energy Dispersive

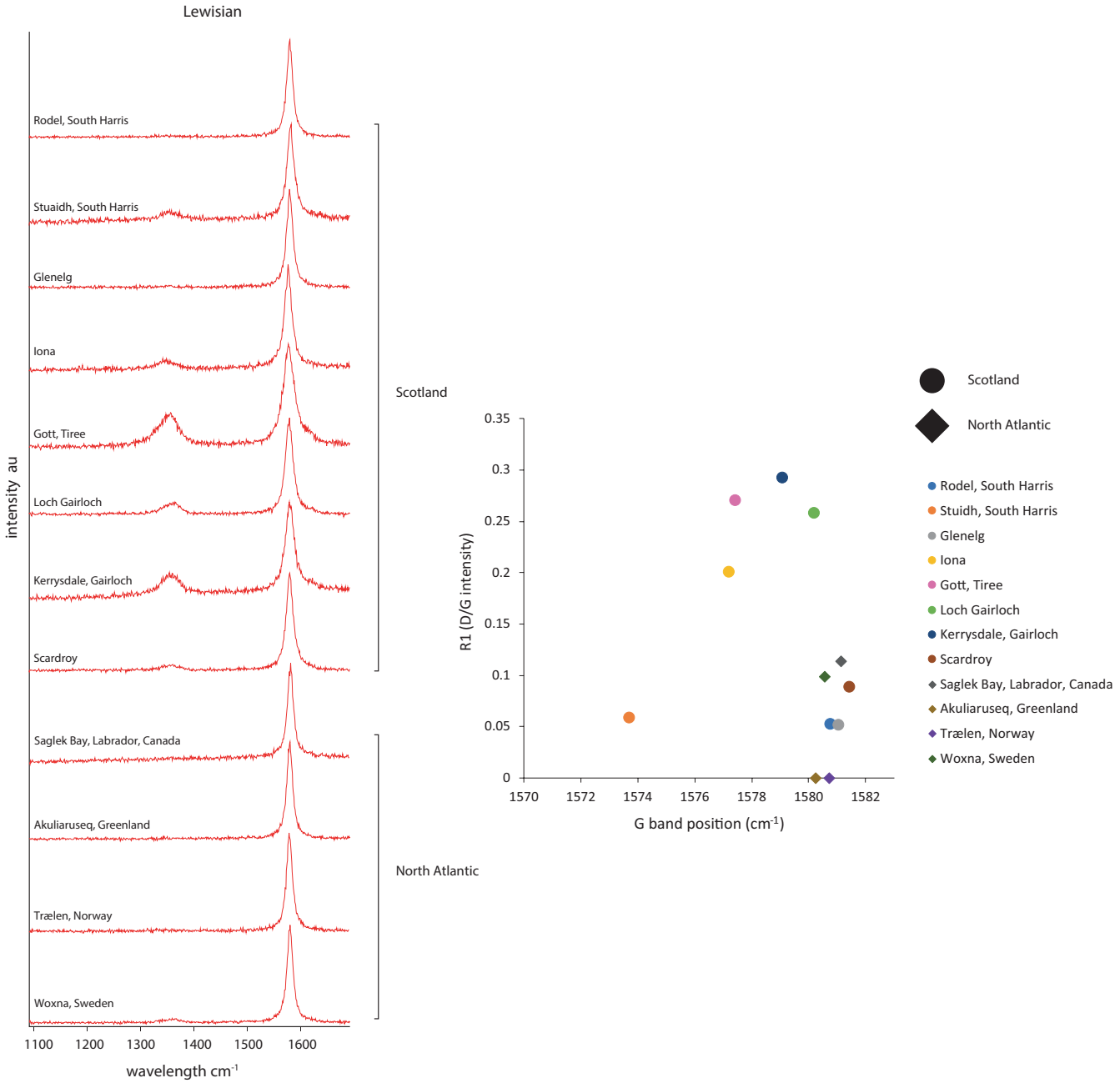

FIGURE 4 Raman spectroscopy spectra for graphite in Lewisian supracrustal rocks and in deposits elsewhere in North Atlantic region. All samples show well-defined graphite order peak (G) at ~1,590 cm^{-1}. Some samples additionally show minor disorder (D) peak at ~1,350 cm^{-1}. Cross-plot of D/G ratio and G peak position emphasizes distinction in samples [Colour figure can be viewed at wileyonlinelibrary.com]

Spectroscopy (EDS) detector, and AZtec software suite. The operating voltage was 8 kV for backscattered analysis (Figure 3).

The structural order of the graphitic samples was characterized by laser Raman spectroscopy, using a Renishaw inVia reflex Raman spectrometer, with a Ar+green laser (wavelength 514.5 nm). The extended spectra in Figures 4 and 5 were based on four spectra each, accumulated over 10 s scan time with 10% laser power. Samples from the Lewisian Complex are compared with samples from active and prospective graphite ore deposits across the North Atlantic region.

Stable carbon isotope analysis was conducted on graphitic samples digested in 10% HCl overnight to remove trace carbonate. Samples were analysed by standard closed-tube combustion method by reaction in vacuo with 2 g of wire form CuO at 800°C overnight. Data (Figure 6) are reported in per mil (‰) using the δ notation versus Vienna Pee Dee Belemnite (V-PDB). Repeat analysis of SUERC's laboratory standard gave $\delta^{13}C$ reproducibility around ±0.2 ‰ (1 s).

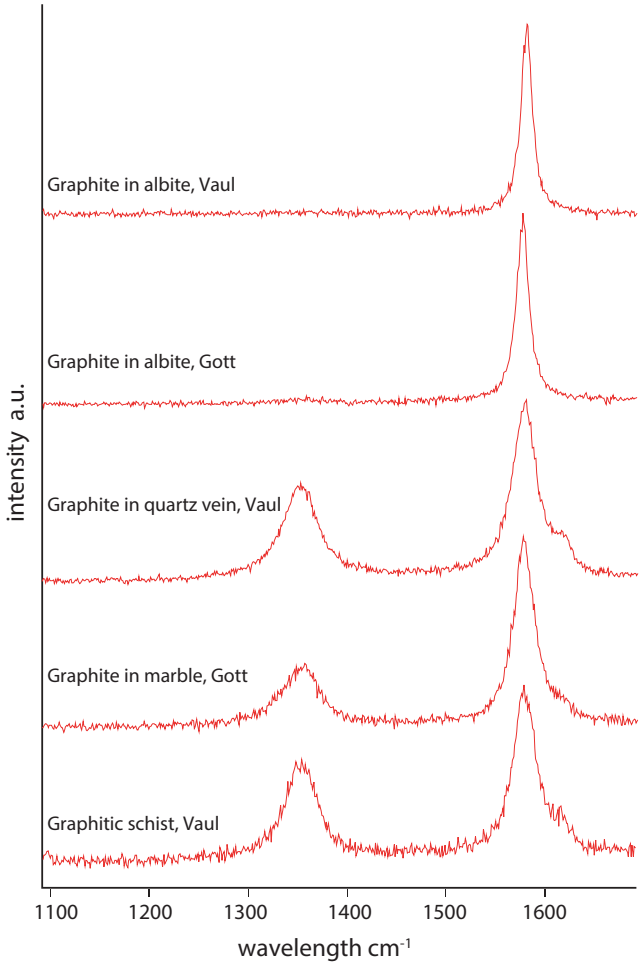

FIGURE 5 Raman spectroscopy spectra for graphite in Lewisian supracrustal rocks from Vaul and Gott, Tiree. All samples show well-defined graphite order peak (G) at ~1,590 cm^{-1}. Samples of marble and schist, and graphitic vein, but not samples in albitized rock, additionally show disorder (D) peak at ~1,350 cm^{-1} and minor disorder (D2) peak as a shoulder on G peak at ~1605 cm^{-1} [Colour figure can be viewed at wileyonlinelibrary.com]

Samples from the Lewisian Complex were supplemented by samples from elsewhere in the North Atlantic region.

4 | RESULTS

The graphite normally occurs as microscopic crystals (less than 0.1 mm crystal size), among quartz, feldspar, mica and other grains in a schistose fabric. In some cases the graphite crystal size is greater, up to 5 mm, conveying a silvery colour to the rock. Graphite accounts for organic carbon contents above 1% (Figure 7). In addition, graphite occurs as partial coatings around phenocrysts, especially allanite, in schists and marbles at Gairloch and Tiree (Figure 3). The phenocrysts are typical of metamorphism in sedimentary rocks, including albite, anorthite, scapolite, dolomite, apatite, allanite and mica (Cartwright, 1992). Albitization is extensive enough to form albitite rock. The metasediments have experienced retrograde metamorphism from 11 kbar and 800°C to greenschist facies (Cartwright, 1992; Westbrook, 1972), but this would not affect graphite, whose structural order is irreversible (Palosaari et al., 2020). At Vaul, Tiree, black graphitic material also occurs in quartz veins cutting the black metasediments.

Graphite occurs in marbles at Tiree and Scardroy. At Gott, Tiree, marble exhibits intermittent 'laminae' of graphite (Figure 2), which are associated with phenocrysts that exhibit rotation. Both 'laminae' and rotation reflect shearing focussed along the marble layers and the supracrustal rocks in general (Westbrook, 1972). At Scardroy, pellets of graphite about 0.1 mm size occur in the marble, especially where the marble is partially replaced by albite. Albitite crystals at Gott and Vaul also contain graphite in vuggy cavities up to 1 mm size.

Representative Raman spectra are shown in Figure 4. Some spectra show a single G peak for ordered carbon. Several spectra additionally show a D peak for disordered carbon. The D peak shows variable degrees of development. It is minor in Iona and Scardroy. Spectra from graphitic beds at two localities in the Loch Maree Group, at Loch Gairloch and Kerrysdale, both exhibit the D peak. A range of samples from Tiree (Figure 5) exhibit the D peak to different degrees, but in the section at Vaul it is strongly developed, and even shows a secondary D2 disorder peak. The G peak positions are typical of graphite (Wopenka & Pasteris, 1993).

The carbon isotope compositions of the graphite have a wide range, which indicates two distinct compositions. Graphite from marble at Tiree and Scardroy has a composition heavier than −10‰, while graphite samples from other localities are lighter than −20‰ (Table 1).

5 | DISCUSSION

Graphite is not abundant in the Lewisian Complex (Cartwright & Barnicoat, 1987), but it is recorded in the supracrustal outliers. A distinct, Palaeoproterozoic, origin for the supracrustal rocks is implied at Gairloch, Glenelg and South Harris, in the range 2.1 to 1.8 Ga. This

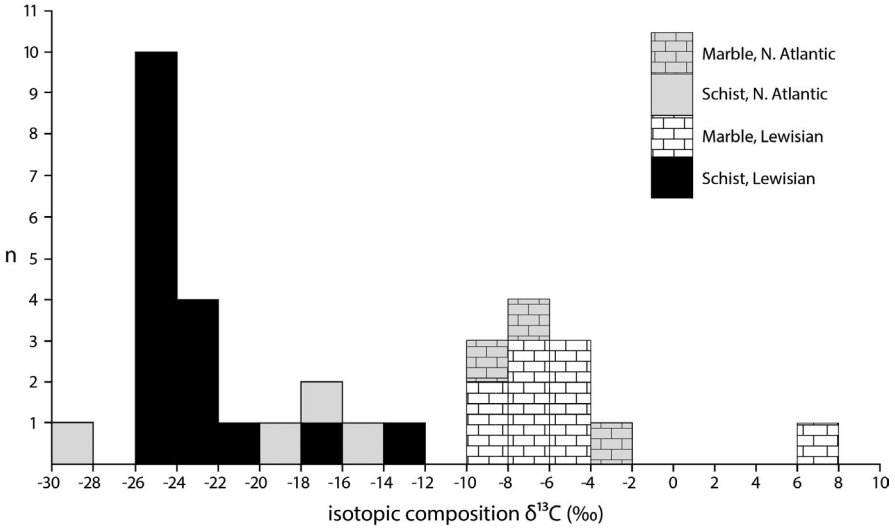

FIGURE 6 Composition of isotopic carbon in samples of graphite in schists and marbles in Lewisian supracrustal inliers, and in selected localities in North Atlantic region (localities recorded in Table 1) [Colour figure can be viewed at wileyonlinelibrary.com]

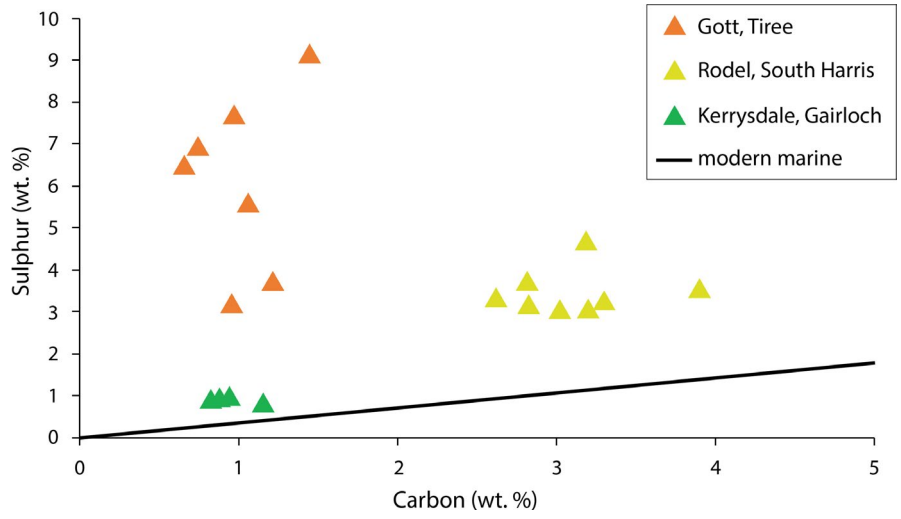

FIGURE 7 Cross-plot of carbon and sulphur contents (wt%) in Lewisian schists, showing sulphur enrichments relative to modern mean ratio due to pyrite formation. Plot after Berner (1982) [Colour figure can be viewed at wileyonlinelibrary.com]

time interval saw graphitic sediments deposited widely in the North Atlantic region, and elsewhere (Condie et al., 2001).

The graphite in schists very probably represents metamorphosed organic matter in the original sediments. However, several occurrences suggest that some graphite was precipitated from a fluid. Vein-hosted graphite at Vaul, Tiree, was clearly a fluid product. The graphite that defines a laminar fabric in marble at Gott, Tiree, formed during shearing and fluid movement during metamorphism. The albitite at Tiree is a replacive and vein-forming rock attributed to metamorphism (Cartwright, 1992), and graphite in cavities in the albitite must have been deposited from a fluid phase. Similarly, the pellets of graphite in albitized marble at Scardroy must also be a fluid product. However, all of the graphite, including that deposited from a fluid, occurs within the supracrustal packages, so have a common origin in the metasediments rather than including a mantle component. It would be Graphite I in the petrographic terminology of Dill et al. (2019).

The Raman spectra for supracrustal rocks show that the carbonaceous material is graphitic rather than kerogenous, based on sharply defined peaks and the position of the order peak (Wopenka & Pasteris, 1993). The spectra for some samples, lacking a D peak, indicate graphite that is fully ordered. However, most samples show at least some degree of disorder. The greatest disorder is exhibited by the samples from Gairloch and most samples from Tiree. Both localities are in amphibolite facies rocks, whereas the other localities are in granulite facies rocks (Table 1). The most fully ordered samples are from South Harris, Glenelg, Scardroy and the albitized rocks of Tiree.

The well-ordered graphite from amphibolite facies albitized rocks at Tiree shows that metamorphic grade is not the sole control on ordering. Previous research concluded that graphite from decarbonation is likely to be fully ordered, while graphite derived from organic matter is less ordered (Pasteris, 1999; Wintsch et al., 1981). Nevertheless, the graphite from marble at Gott shows disorder (Figure 5). Although there is not a simple relationship between ordering and evidence for deposition from fluid in the Lewisian Complex, most of the fluid-derived samples are well-ordered.

TABLE 1 Isotopic composition of graphite samples

Inlier	Locality	Grid ref.	Lab number	Setting (met. grade)	$\delta^{13}C$ (‰)
Lewisian Complex					
Gairloch	Kerrysdale	NG 822736	PPG6	Schist (Amphibolite)	−24.0
Gairloch	Kerrysdale	NG 822736	PPG7	Schist (Amphibolite)	−24.5
Gairloch	Kerrysdale	NG 822736	PPG8	Schist (Amphibolite)	−24.4
Gairloch	Kerrysdale	NG 822736	PPG9	Schist (Amphibolite)	−24.4
Gairloch	Kerrysdale	NG 822736	PPG26	Schist (Amphibolite)	−23.6
Glenelg	Sgiath Bheinn	NG 8218	PPG47	Schist (Granulite)	−21.0
Glenelg	Sgiath Bheinn	NG 8218	PPG21	Schist (Granulite)	−22.6
Glenelg	Sgiath Bheinn	NG 8218	PPG22	Schist (Granulite)	−22.6
South Harris	Rodel Pier	NG 047830	PPG13	Schist (Granulite)	−24.2
South Harris	Rodel Church	NG 048832	PPG15	Schist (Granulite)	−25.1
South Harris	Rodel Church	NG 048832	PPG16	Schist (Granulite)	−24.9
South Harris	Stuaidh	NG 043832	PPG10	Schist (Granulite)	−24.5
South Harris	Stuaidh	NG 043832	PPG11	Schist (Granulite)	−24.8
South Harris	Stuaidh	NG 043832	PPG12	Schist (Granulite)	−25.0
Tiree	Vaul	NM 050488	PPG25	Schist (Amphibolite)	−17.4
Tiree	Gott	NM 045456	PPG54	Marble (Amphibolite)	−7.5
Tiree	Gott	NM 045456	PPG55	Marble (Amphibolite)	−8.4
Tiree	Gott	NM 045456	PPG55A	Marble (Amphibolite)	−6.7
Tiree	Gott	NM 045456	PPG59	Marble (Amphibolite)	−8.9
Tiree	Gott	NM 045456	PPG66	Albitized gneiss (Amp)	−13.2
Tiree	Gott	NM 045456	PPG69	Marble (Amphibolite)	−7.3
Tiree	Gott	NM 045456	PPG69A	Marble (Amphibolite)	−5.9
Iona	NW Iona	NM 263248	PPG1	Schist (Granulite)	−23.3
Scardroy	Scardroy	NH 223523	PPG49	Marble (Granulite)	−5.1
Scardroy	Scardroy	NH 223523	PPG63	Marble (Granulite)	−4.0
Scardroy	Scardroy	NH 223523	PPG 64	Marble (Granulite)	+6.2

Country	Locality	Lab number	Setting	$\delta^{13}C$ (‰)
North Atlantic Region				
Canada	Saglek Bay, Labrador	PPG76	Schist	−29.83
Canada	Soper River, Kimmirut	PPG74	Marble	−8.65
Canada	Soper River, Kimmirut	PPG75	Marble	−7.74
Greenland	Akuliaruseq	PPG80	Schist	−15.22
Norway	Traelen, Skaland	PPG4	Schist	−18.7
Sweden	Woxna	PPG48	Schist	−17.7
Finland	Skrabbole, Pargas	PPG70	Marble	−2.60

Graphite in sediments from the 2–1–1.8 Ga interval elsewhere, from Greenland, Norway, India and Argentina, similarly exhibits disorder despite metamorphism to granulite and amphibolite facies (Lajoinie et al., 2015; Mishra & Bernhardt, 2009; Palosaari et al., 2016; Papineau et al., 2009; Rosing-Schow et al., 2017). In contrast, graphite veins, concentrated from Palaeoproterozoic sediments so they can be exploited as graphite ore deposits, typically show complete ordering. High ordering is evident in the mined and prospective deposits in Labrador, Greenland, Norway and Sweden (Figure 4). These data, showing high ordering in graphite deposited from fluid, are consistent with the data for graphite in Scotland.

The graphite with isotopic composition in the range −20 to −30‰ is typical of graphite derived from organic matter. The successions containing graphite within this range of compositions, in South Harris, Gairloch, Glenelg and Iona, are all schists that were probably deposited in low-energy marine environments. With two exceptions, the values in schists are more precisely all in the range −21 to −25‰, which is typical of other Palaeoproterozoic

graphitic carbon. For example, coeval datasets from Greenland, Australia and China schists have mean values of −24.5‰ (n = 11), −24.1‰ (n = 11) and −24.2‰ (n = 12) respectively (Rosing-Schow et al., 2017; Williams, 2007; Zhong et al., 2019). The distinct composition of the graphite in marbles (heavier than −10‰, mostly −4 to −9‰) is similar to that derived from magmatic carbon dioxide (Luque et al., 2012) However, it can be explained by a source of carbon dioxide in decarbonation of marbles with a near-zero composition. Similar combinations of graphite with light compositions in pelites and heavier compositions near marbles, in amphibolite and granulite facies, are reported by Weis et al. (1981) and Baker (1988). The two exceptions to the tight range of values for the schists may represent mixing of organic matter and carbonate sources.

Marbles of Palaeoproterozoic age across the northern hemisphere are consistently associated with graphite, from Baffin Island, Canada (Belley et al., 2017) to West Greenland (Garde, 1978), Finland (Lehtinen, 2015), Tajikistan (Sorokina et al., 2015) and China (Yang et al., 2019). We have measured the carbon isotope composition of graphite in marble from Kimmirut, Baffin Island, and Pargas, Finland, at −8.1 and −2.6‰ respectively (Figure 6). These heavy compositions indicate, like the graphite in marble from Scotland, an origin in decarbonation of the marble. We note that the Palaeoproterozoic graphitic marbles in Baffin Island, Tajikistan and China all host gem quality corundum (ruby and sapphire).

A further aspect of Palaeoproterozoic graphite is a consistent occurrence in albitite veins, as in Scotland. Examples include deposits in Brazil (Sirqueira et al., 2018), Russia (Sorokhtina et al., 2010) and India (Mukherjee et al., 2016), which emphasize graphite precipitation from fluids was widespread.

6 | CONCLUSIONS

Graphite occurs in numerous supracrustal successions within the Lewisian Complex. Petrographic, isotopic and spectroscopic studies show that:

(i) The graphite in schists is derived from sedimentary organic matter, while graphite in marbles is derived from decarbonation of limestone.
(ii) In addition, there is evidence of graphite deposition from migrated (i.e. fluid) carbon, in cross-cutting veins and cavities.
(iii) Some of the graphite is fully ordered, especially where it was deposited from a fluid. However, much of the graphite is not completely ordered. Graphite examined from Gairloch and Vaul, Tiree, shows disorder, despite metamorphism to amphibolite facies.
(iv) Comparison of the data from Scottish graphite with that of exploitable deposits in the North Atlantic region shows that ordered graphite that may be commercially valuable is more likely to occur in veins, which should guide exploration.

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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