Alteration fabrics and mineralogy as provenance indicators

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Supplementary information

Petrographic description of spotted dolerite sample CMQ2 from Carn Meini

The surface of dolerite CMQ2 is greenish-grey (5G 5/1 on the Geological Society of America rock-color chart) and carries white spots with thin, <1 mm wide, greener rims. The cut surface is a light bluish-grey (5B 7/1) with 10 mm diameter, bluish-white (5B 9/1) spots.

Transmitted light microscope examination shows large plagioclase laths (An_{44}) that are zoned, twinned and extensively altered to fine-grained clinozoisite with very low interference colours and white mica; epidote with high interference colours and clinochlore also replace plagioclase. The margins of this generation of plagioclase are often unaltered. A later generation of smaller, stubby plagioclase (albite?) is associated with chlorite. The main generation of plagioclase has a sub-ophitic relationship with clinopyroxene. Small plagioclase laths enclosed in titanomagnetite are less altered than the main plagioclase. Most clinopyroxene, some of which is zoned or twinned, is unaltered, as are acicular apatite crystals, some of which are associated with later stage plagioclase (albite?).

Secondary minerals are abundant and include late-stage quartz, with some quartz forming spherulitic aggregates or thin, cross-cutting veinlets. Zoned epidote commonly occurs as mosaics and in epidote-chlorite (clinochlore with brown interference colours)-pyrite intergrowths, some of which surround clinopyroxene. Unpublished electron microprobe analyses by one of us (REB) record a range in XFe values in Preseli dolerites ranging from 0.11 to 0.33 (where XFe = $Fe^{3+}/(Fe^{3+}+Al)$), spanning the clinozoisite-epidote boundary. Our unpublished data show that sample PCC11 from Carn Goedog has XFe values of 0.29 (core) and 0.15 (rim) respectively. This zoning reflects a decrease in Fe contents towards the crystal rims, a feature which Grapes and Hoskin (2004) interpreted as resulting from prograde metamorphism conditions during crystal growth. Within the chlorite-epidote intergrowths euhedral epidote is enclosed within chlorite or forms euhedral rims to chlorite infills. Where these intergrowths are rounded and occur in clinopyroxene crystals they have been interpreted by Bevins et al. (1989) as being pseudomorphs after olivine. Minor amounts of chamosite are present. Very locally, small, 20-30 μm long, rhombic titanite is enclosed within chlorite. Green to yellow-green, fibrous fringes with moderate relief and showing high interference colours are identified as amphibole and have been shown by

Bevins and Rowbotham (1983) on the basis of electron microprobe analysis to be actinolite; they grow out from clinopyroxene or epidote and grow into quartz or chlorite. Minor amounts of titanite mantle highly altered ilmenite.

The mineralogy of the 'spot' areas are dominated by masses of very small crystals. Because of their fine grain size the crystals appear to be isotropic due to grain boundary refraction effects. They are very altered primary plagioclase crystals which now comprise abundant, fine-grained clinozoisite with very low interference colours intergrown with white mica; they also carry chrome spinel. Epidote with high interference colours, chlorite (clinochlore) lying along relict cleavage and relict feldspar/secondary feldspar are less abundant. Locally white mica is the main alteration product.

Reflected light microscope examination shows that the chrome-rich spinel forms rounded grains, 40-80 but up to 250x200 μm in size with brown internal reflections and thin, pale 'ferrochromit' veinlets or pitted, spongy, altered rims. Ixer (in Darvill et al., 2009), using reflected light microscopy, reported the presence of small (10-40 but up to 80 μm diameter) euhedral crystals or 150-250 μm diameter 'pitted' rounded grains of chrome spinel in spotted dolerite CM2 from Carn Meini; similar pitted, rounded grains are reported in PCC11 from Carn Goedog. Ixer (op. cit.) suggested that they were restricted to the spot areas but subsequently we have observed them also in clinopyroxene crystals. Ixer (in Thorpe et al., 1991) reported the presence of 200 μm diameter spinel in a spot in Stonehenge orthostat SH61. Analyses of euhedral chrome spinels show they are either spinel or chromite, following the classification of the spinel supergroup by Bosie et al. (2019). Semi-quantitatve analyses of spongey spinels from sample PCC11 show that they are depleted in magnesium at the expense of the introduction of zinc.

Large, equant crystals of titanomagnetite are extensively altered. Magnetite has altered totally to fine-grained titanite with white internal reflections, whilst crystallographically orientated, 1-2 μm wide ilmenite oxidation exsolution lamellae have altered to fine-grained, acicular, colourless TiO_2 minerals or, if thicker (between 2-5 μm wide), to TiO_2 minerals with orange internal reflections. Titanomagnetite carries rare, 10 μm diameter pyrite or limonite pseudomorphs after pyrite. Ilmenite, 80-100 μm long, is intergrown with titanomagnetite as an internal or external sandwich and is altered to pale-coloured TiO_2. Elsewhere, much ilmenite forms discrete, lobate laths up to 200 μm in length, or symplectite-like intergrowths with silicates. All generations of ilmenite are altered extensively. Some ilmenite alters to orange TiO_2 but most is replaced by fine-grained mixtures of 5-10 μm diameter, bireflecting, colourless probable carbonate and small, 2-5 but up to 10x2 μm long, pale-coloured to yellow TiO_2 minerals. The latter are crystallographically controlled with respect to the original ilmenite grain. The standard alteration sequence is ilmenite to 'pitted/spotty' ilmenite to carbonate to TiO_2 minerals and finally to titanite. Relict ilmenite up to 10-20 μm in diameter is commonly present. Wispy titanite rims up to 100 μm in thickness enclose ilmenite.

Sulphides are present in minor to trace amounts. Pyrite is the most abundant, forming 10-200 μm diameter, euhedral crystals, locally collected into aggregates. Rarely pyrite encloses 2-5 μm diameter, mixed chalcopyrite-pyrrhotite inclusions. Much pyrite has oxidised to limonite. Very rare, 10 μm diameter pyrite is enclosed within altered titanomagnetite but most surrounds titanomagnetite.

Pyrrhotite, as 20-40 μm diameter, hexagonal grains, 5-60 μm diameter chalcopyrite (or their limonite pseudomorphs) and 10-20 μm diameter, mixed chalcopyrite-pyrrhotite grains occur within epidote. Trace amounts of 10-40 μm diameter pyrrhotite have altered to limonite and patches up to 200 μm in diameter of 2-15 μm diameter chalcopyrite or 2-5 μm diameter pyrrhotite are present in alteration minerals.

References

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Geological terminology definitions

Magma

Hot fluid or semi-fluid material below or within the Earth's crust. When magma is erupted at the Earth's surface it produces lava flows or ash eruptions. If the magma rises but is arrested at a high level in the crust it forms shallow-level intrusions, as in the dolerites of the Mynydd Preseli.

Primary mineral

The primary minerals in the dolerites are those which crystallize in magma when it is intruded into the Earth's crust some 450 million years ago. They crystallize at temperatures in the range between ca. 1300C to ca. 800C. In the Preseli dolerites the primary minerals are olivine, clinopyroxene, Ca plagioclase feldspar, apatite, 'spinel' and an iron-titanium oxide. All of these minerals are anhydrous (ie they contain no water in their crystal structures).

Secondary mineral

Following crystallization the high temperature primary minerals they are potentially unstable at the relatively low temperatures of the Earth's crust and are subject to breakdown (alteration), especially in the presence of circulating fluids which act as a catalyst for recrystallization. A range of secondary minerals developed in the Preseli dolerites, including albite, chlorite, zoisite, clinozoisite, epidote, actinolite, titanite, prehnite and pumpellyite. Many of these minerals are hydrous (ie they have water in their crystal structures).

Phenocryst/glomerocryst

As molten magma cools crystals grow in the melt early formed crystals have the opportunity (space) to grow to quite large sizes. This typically takes place where magma is held in the Earth's crust in a magma chamber. When magma rises to erupt at the Earth's surface or is intruded into relatively cold crust the remaining melt crystallizes leading to the generation of a mass of smaller crystals, effectively freezing in the larger, early formed crystals. These early formed crystals are called phenocrysts or if they formed in aggregates of crystals they are called glomerocrysts.

Saussuritization

Saussuritization is the process of alteration of early-formed, high temperature calcium-rich plagioclase feldspar. The alteration commonly relates to the circulation of late magmatic fluids through the cooling rock body. The primary mineral is replaced by a secondary mineral assemblage which includes albite, zoisite, clinozoisite and muscovite.