The bloomery mounds of the Scottish Highlands. Part 2: A review of iron mineralization

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Abstract

The regional and local geology underlies the availability and type of natural resources, be it ore, woodland or water, and was the fundamental influence on the locations chosen for iron making. Possible sources of iron ore available to early bloomery iron workers in Scotland have been established from the recorded sources of iron ore. Moreover, using modern knowledge of iron geochemistry as well as the local and regional geology, further potential sources of iron may be predicted. The question of the original ore used at a bloomery site can be tackled by looking for the remains of ore processing/crushing, using techniques such as ore petrography. In spite of much geological evidence for potential iron ores other than bog ore, no positive evidence for their use was found at the bloomery sites investigated. Iron-rich manganiferous slags with minor silica suggest that bog iron ore was used.

Introduction

Research into bloomery iron production in Scotland has for long lagged behind similar studies in England and Wales (Tylecote 1986). Apart from the extensive early surveys by Macadam (1887) of bloomery mounds in the Highlands and Lowlands of Scotland, and of Dixon (1886), in the parish of Gairloch, Ross-shire, little interest had been shown in their recording and study until the late 1960s when Aitken (1970) surveyed and partly excavated bloomery mounds in Perthshire and Argyllshire. Yet these 'monuments' are an ubiquitous feature of the Scottish landscape. Because of their frequent occurrence, it was inevitable that they would fall prey to land reclamation for agricultural purposes, to forestry ploughing and to animal burrowing. In this way, important evidence relating to the early Scottish iron industry and the social and economic factors that sustained it has been slowly and inexorably vanishing. This critical state of affairs prompted a one-year pilot investigation of some of these monuments and potential means of preserving them. Part I of this investigation (Photos-Jones *et al* 1998) highlighted various bloomery iron-making practices, from the traditional to the most technologically advanced, during a period from the late medieval to the early 17th century. Two areas, the Cowal Peninsula in Argyll and Loch Maree in Western Ross (Fig 1) were chosen to highlight three stages of bloomery iron-making, from the simple one-man type of operation to the clan-type organization, and finally to that of an industrial complex based on imported technological know-how. In all cases, the regional and local geology, underlying the availability and type of natural resources, be it ore, woodland or water, was seen to influence the location chosen for iron making. This report aims to highlight the nature of the natural earth resources, with particular emphasis on the diverse types of iron-ore occurrences in Scotland. It also attempts a more precise characterization of bog iron ore in the Highlands, a term long overused in the archaeometallurgical literature, but

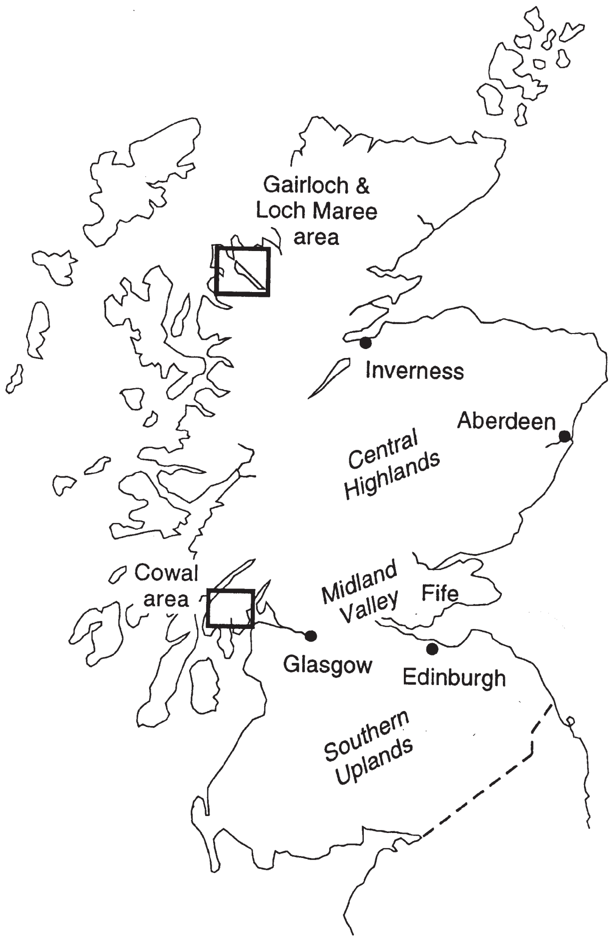

Figure 1: Main study areas of the investigation of bloomery mounds; see Photos-Jones et al *1998 for more detailed maps.*

little understood. The reason has been the complex geochemical factors associated with its formation and regeneration, and the lack of appropriate ethnographic evidence.

A geological contribution to the archaeological investigation of bloomery iron-making was initiated primarily to ascertain whether a geological/prospecting approach could assist in the determination of the nature and source of iron ores used at bloomery sites. This geological contribution is divided into two sections. The first provides a discussion of prospecting for iron ores and a brief account of iron ore minerals. The second reviews iron ores with emphasis on those iron ore deposits of Scotland of relevance to the bloomery investigation; this incorporates illustrative case studies from the areas selected for the archaeological study.

Prospecting

Prospecting can be considered in two ways: the possible prospecting methods used by the early iron workers, and the modern approach used to help establish sources of iron ore formerly in use.

Early prospecting

We can only speculate on the methods used by early Highland prospectors. Once the characteristics of iron ore deposits were known to them, it is likely that they explored by following streams in an upstream direction, starting from convenient access points, and looking for visible clues. An identical approach is still used today, but supported by a large database of background geological information. Even without a detailed understanding of geological processes, it would have been possible, on the basis of experience, to use the following evidence to help locate iron ore deposits: firstly, the nature of the terrain (*eg* poorly drained flat-lying, or steeply incised valley); secondly, boulders in stream floors (*eg* fragments of iron ore, or brown-stained rocks); thirdly, outcrops in streams; and finally, reddish-brown seepages into streams (*ie* from tributaries or from fractures in outcropping rock), for which the Gaelic term is ‘meinn’.

The modern approach

The starting point for a modern assessment of possible sources of iron ore available to the early bloomery iron workers is to establish the known sources of iron ore in Scotland, recorded in the geological literature. Scotland had a long history of iron ore production but there are now no economically viable deposits. The deposits can be quite complex in detail, so iron ore mineralogy, geochemistry and mineralization processes must be considered. Together with knowledge of the local and regional geology, further potential sources of iron may be predicted. The pilot investigation of bloomery sites (Photos-Jones *et al* 1998), established the following four stages for the geological investigations:

The regional geological setting obtained from textbooks and maps provides an indication of the potential for different types of iron ore deposits. Iron ore deposits and possible workings may be recorded nearby or in the same geological terrain. This also provides a basis for fieldwork and consideration of the local geology.

The nature of the geology within a kilometre or so of a bloomery site can be assessed from maps and supplemented by fieldwork. Closer to a bloomery, the best approach is to traverse the area, especially by using nearby streams where outcrops and boulders reveal the local rock types.

Clues to iron mineralization are often provided by boulders found in streams. This is because streams are often located where rocks are fractured and mineralized. Boulders containing iron-mineralization can be traced to source. In the same way, bloomery slag found in a stream may be followed upstream to where the site is being eroded. Iron-rich seepages can also reveal iron-enriched rocks. However, brown iron-rich coatings on boulders and seepages into streams are not uncommon, and some care and skill is required in recognizing whether iron-enrichments may be related to economically-significant mineralization.

The question of the original ore used at the site can be tackled by looking for the remains of ore processing/crushing. Concentrates of heavy minerals can be recovered by panning, using soil which is suspected of containing ore (Allt na Ceardaich, Photos-Jones *et al* 1998, 22). The bulk mineralogy of the concentrates can be determined using X-ray diffraction spectrometry while polished thin-section petrography using transmitted and reflected-light microscopy can be used to identify ores and smelting products in grains of about 100 micrometre size. Petrographic study of the same samples can be aided by using scanning electron microscopy with elemental analyses. Because of the potential textural complexity of variably heated minerals and metallurgical products, this promising approach requires the development of special expertise in order to interpret such samples.

The major iron ore minerals

Reference to iron ores in relation to prospecting for ore deposits and iron smelting requires an understanding of the mineralogy of iron ores, and we make frequent reference to these. The mineralogy of the major iron ore minerals is therefore summarized below:

Magnetite (Fe_3O_4) is a black magnetic mineral found as a minor component in many igneous and metamorphic

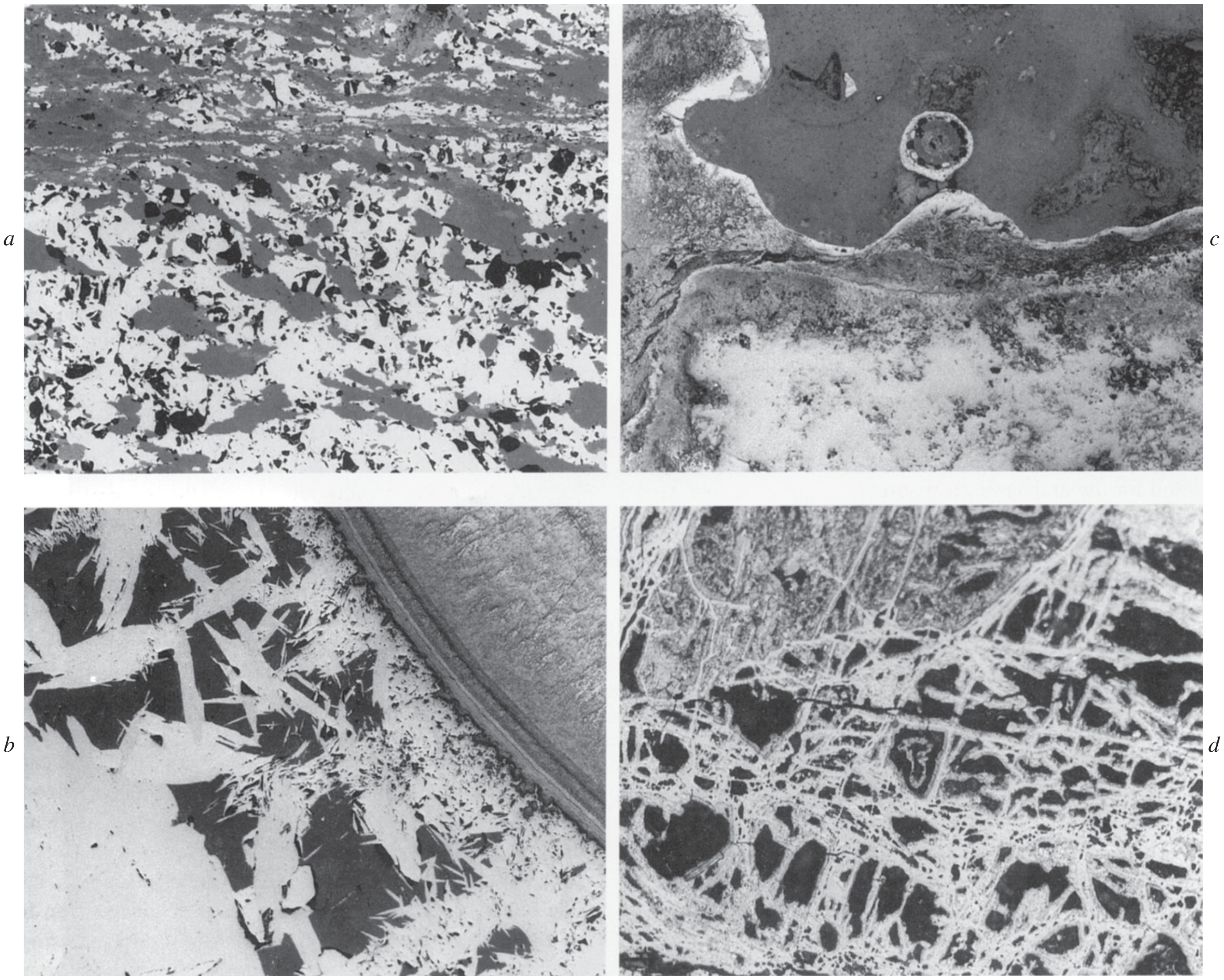

Figure 2: Polished sections. a) Typical magnetite schist, Gairloch area (4mm across). The magnetite is light grey. Note the distinctive preferred orientation of the silicates (grey), typical of a metamorphic rock. b) Typical kidney iron ore of the Cumberland type, Muirkirk, Southern Uplands (4mm across). Platy to bladed hematite crystals (white) set in quartz (dark grey) form a zone between thinly layered cryptocrystalline hematite and quartz, and coarse grained massive hematite. c) Bog iron ore from the iron dyke, South Erradale, near Gairloch (2mm across). The goethite is light grey and tends to be brighter as it becomes more crystalline and/or manganiferous. A cross section of a plant stem is seen near the centre of the field at the edge of a large cavity (dark grey). Micro-inclusions (dark) of mineral and plant debris are widespread. d) Goethite (light grey) from a gossan showing the boxwork texture; cavites are black (2mm across). This distinctive texture results from oxidation of pyrite.

rocks. Following erosion and weathering of magnetite-bearing rocks, it can accumulate as a detrital and chemically-resistant heavy mineral in sediments, for example in black beach sands as found locally on the west coast of Scotland. Such magnetite is usually titaniferous and intergrown with Fe-Ti-O minerals, eg ilmenite ($FeTiO_3$), rutile (TiO_2) and hematite (Fe_2O_3). Magnetite can be quite impure, containing manganese, magnesium, vanadium, aluminium and/or chromium in solid solution. Magnetite skarn deposits are pods and veins of replacive magnetite found in limestones adjacent to granites, and such a deposit is found in Skye. Some metamorphosed sedimentary rocks may also be magnetite-rich. MacGregor *et al* (1920) report that the magnetite-bearing schists (Fig 2a) in the Loch Maree area are of too low a grade to be used as ore; however many factors influence economic viability, so such magnetite is a potential ore for the Fasagh site, Loch Maree (Photos-Jones *et al* 1998, 24, 28). Magnetite is usually variably oxidized to hematite, and in polished section closely resembles oxidized wustite (FeO) from bloomery slag. Textural relationships are invaluable for distinguishing magnetite from wustite, which is very rare in nature.

Hematite (Fe_2O_3) can be found as a minor component in virtually any rock type, but concentrations of hematite are typically in the form of veins of hydrothermal origin. There is a good example at Muirkirk in the Southern Uplands, where the vein contains massive botryoidal ore (Fig 2b) very similar to that produced from the well known Cumberland deposits (Shepherd and Goldring 1993). Quartz (SiO_2), calcite ($CaCO_3$) and/or baryte ($BaSO_4$) commonly accompany hematite, but hematite itself is usually a very pure mineral. Reddish-brown coloured rocks are often said to be 'hematitic' but the reddish colour can be imparted by a very small amount of micron-sized hematite which transmits red light. Crystalline hematite is opaque, metallic black, and often platy.

Iron oxyhydroxide is the general name (Waychunas 1991) for amorphous and crystalline hydrous precipitates of mixed-valence iron oxides (Fe^{2+} and Fe^{3+}). FeOOH (with ferric iron only) is found as two types of crystalline structures (polymorphs). These are the minerals goethite and lepidocrocite. Ferrihydrite is a poorly crystalline substance close to $5Fe_2O_3.9H_2O$ in composition. The brown weathering product from the ferrous iron-bearing minerals found on many rocks is commonly called 'limonite'. This is a poorly crystallized mixture of iron oxyhydroxides corresponding to $FeOOH.nH_2O$ (Oades 1963).

Progressive dehydration and/or heating of ferric hydroxide precipitates leads to increasing crystallinity and to the formation of goethite followed by hematite (Fischer and Schwertmann 1975). This is an important consideration when investigating potential bog-iron ore at a bloomery site and the three stages can be 'fingerprinted' using X-ray diffraction (see Figure 3). Hydrohematite and protohematite are intermediate phases formed on transition of FeOOH to Fe_2O_3 (Waychunas 1991). Limonite can be yellow, brown or black but becomes redder if it recrystallizes to fine hematite, Fe_2O_3. Mixed ferrous/ferric iron-bearing hydroxides are probably rare in nature but could in theory dehydrate to give magnetite with hematite. Ferrous hydroxides are unlikely to be found in nature.

Bog iron ores consist of ferric oxyhydroxides (Fig 2c) and are probably widespread in Scotland. We are not aware of any review of their occurrence. They are potential ores for all the bloomery sites investigated in this project.

Siderite ($FeCO_3$) is a grey- to brown-coloured mineral, becoming dark brown as it oxidizes to goethite which encrusts and replaces the siderite. It often contains Mg, Mn and Ca in solid solution. It is found in veins, in chalybeate spring deposits, as concretions in clayband ironstones, and as beds in blackband ironstones. It can also be found in bog iron ores (Postma 1977).

Pyrite (FeS_2) can be enriched in some igneous rocks and hydrothermal veins, and thick layers of hydrothermal pyrite can form in the sedimentary environment. On metamorphism such pyritic layers can transform, by loss of sulphur, to pyrrhotite, $Fe_{1-x}S$. Both pyrite and pyrrhotite are prone to oxidation, to give secondary deposits of limonite/hematite. Iron sulphide-rich metamorphic rocks are quite common in the Highlands (Hall 1993) and are likely to be source rocks for small gossanous and bog iron ore-type deposits, especially in the Loch Maree area (see below). Goethite after pyrite has a distinctive boxwork texture (Fig 2d) and relict inclusions of pyrite in gossans might explain why inclusion of iron sulphides have been found in some iron slags. However, pyrite from coal is an alternative source.

Chamosite, a 2:1 trioctahedral chlorite hydroxy iron silicate ($Fe_3Al_2Si_2O_{10}.3H_2O$), can form in unusual iron-rich sedimentary environments leading to beds of sedimentary iron ore. These are often oolitic (a characteristic of the so-called Minette or Clinton-type iron ores) and the iron can be variably oxidized to goethite/hematite. The Raasay ironstone of Jurassic age is an example and was discovered and worked at the beginning of the 20th century (MacGregor *et al* 1920).

Iron ore deposits of Scotland

Scottish iron ore deposits have been reviewed extensively by the Geological Survey in special reports such as that by MacGregor *et al* (1920), who classified the ores as: bog iron ores of Recent origin; hematite ores of different ages; Carboniferous clayband ores; Carboniferous blackband ores; and Jurassic ores.

The Survey's review concentrates on the major iron ore deposits which were exploited primarily in the last two hundred years. None are now viable. There is additional information scattered through the geological literature and on Geological Survey maps. It is noteworthy that small iron ore deposits, in particular bog iron ores, could well remain unrecorded. Other small deposits could include magnetite sands, chalybeate springs and siderite veins (possibly oxidized). A recent review of sedimentary iron ore in the UK is given by Young (1993) .

The present review concentrates on the nature and origin of different types of iron deposit which have a bearing on the bloomery investigation. These include bog iron ores, replacement encrustations, hematite, clayband and blackband ironstones and magnetite schists.

Bog iron ores

Bog iron ores have long been considered as the most significant source of iron for the early iron smelters. Bog

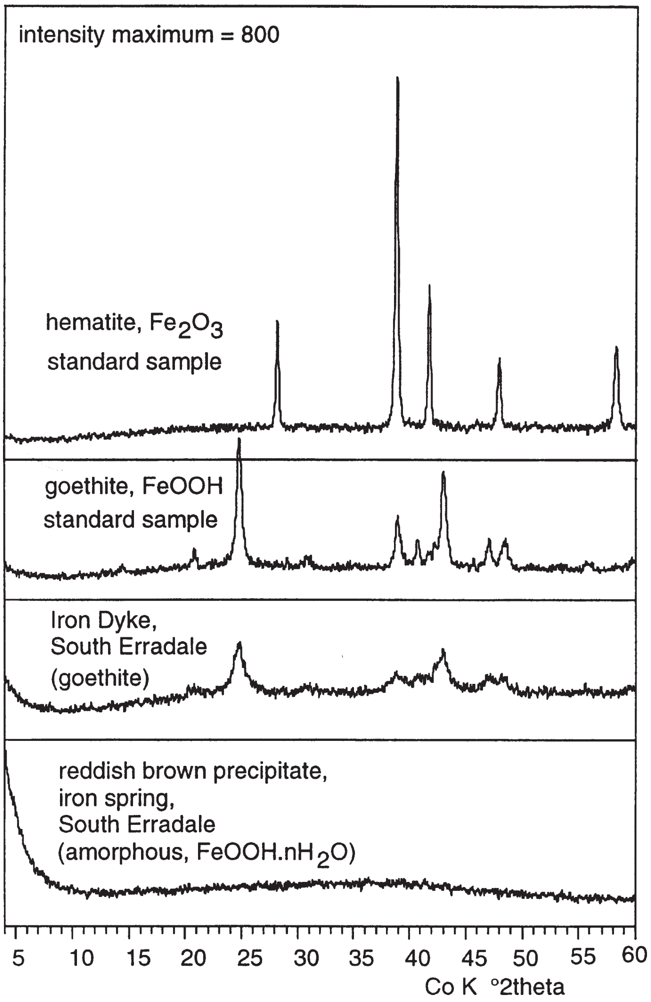

Figure 3: Powder X-ray diffraction profiles of representative samples of iron oxyhydroxides to demonstrate the progressive increase in crystallinity from amorphous ferric iron precipitates through to hematite. Increase in crystallinity is indicated by increase in sharpness of the peaks which represent interlayer spacings in the atomic structures of the minerals. Goethite and hematite have different atomic structures so the peaks are in different positions.

iron ores are accumulations of ferric oxyhydroxides with the general formula $FeOOH.nH_2O$. When pure, bog iron ore contains 60 wt% iron (Winchell 1891). These have precipitated where acidic iron-bearing groundwaters have been oxidized and neutralized on seeping into oxidizing surface or near-surface environments (Fig 4). Such seepages are very common. However, a productive source of iron, an aqueous environment amenable to iron dissolution and an effective precipitation mechanism, would be necessary to produce a useable quantity of iron ore.

Ferric iron (oxidized iron, Fe^{3+}) is known to be exceedingly insoluble except in extremely acidic water, while ferrous iron (reduced iron, Fe^{2+}) is readily soluble and can be transported in solution provided the solution is isolated from oxygen. This is evident on an Eh-pH diagram (Fig 4), which shows the degree of oxidation and the pH of common natural waters near the surface of the Earth.

There are many rocks which contain iron-rich minerals which may weather to produce acidic solutions with dissolved ferrous and ferric iron. Some acidity is provided by rainwater, but decaying organic matter and/or the oxidation of sulphide minerals to sulphuric acid can greatly increase acidity and solubility of iron complexes. Ferric-iron-reducing bacteria contribute to the process (Brock *et al* 1994). Bleaching of rock under peaty soils is a typical result of iron dissolution. A major reason for precipitation of iron is oxidation of soluble ferrous cations and complexes to insoluble ferric cations and this takes place where groundwaters come to the surface. Iron invariably precipitates as amorphous ferric iron oxyhydroxides. Goethite is used on the computed Eh-pH diagram to represent precipitated ferric iron in general. However, it is now well established that bacteria are often involved in this process. Bacteria can enhance iron precipitation in several ways. Iron bacteria (Brock *et al* 1994) use ferrous iron as an energy source and oxygen as an electron acceptor, and thus cause the conversion of ferrous to ferric iron. Some iron bacteria which take advantage of the acidic

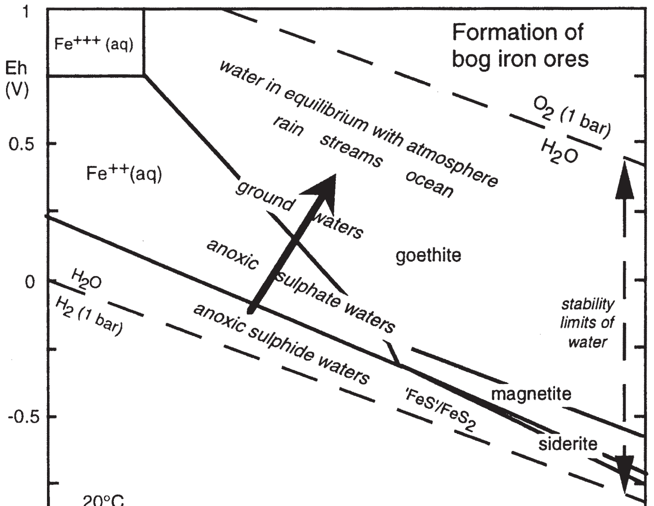

Figure 4: Natural waters and stability field of iron minerals shown on an Eh-pH diagram computed using Geochemist's Workbench (Bethke 1996) for nominal activities of $Fe^{2+}(aq) = 10^{-6}$, $SO_4^{2-} = 10^{-10}$ and fugacity of $CO_2 = 10^{-3.5}$ (atmospheric). Magnetite (Fe_3O_4) and siderite ($FeCO_3$) fields shown. Hematite (Fe_2O_3) suppressed to show goethite (FeOOH) which is a more realistic control on iron solubility at low temperature. Pyrite (FeS_2) is thermodynamically stable but 'FeS' phases are favoured kinetically. The arrow indicates how ferrous iron, soluble in reduced (anoxic) acid water, would be precipitated as goethite with increase in oxidation. Siderite is only stable in anoxic environments at atmospheric or higher carbon dioxide partial pressures. Siderite and iron sulphides are common in sediments. Magnetite however is rare, probably because of kinetic inhibition of nucleation.

conditions to enhance their energy transfer mechanism, *eg Thiobacillus ferroxidans*, use ferrous iron in very acidic waters such as those draining oxidizing iron sulphide deposits. Others, such as *Gallionella ferruginea*, live in near-neutral waters at the interface of anoxic and oxic conditions. The bacteria have to precipitate a great deal of iron to obtain a useful energy supply. Photosynthetic bacteria and/or algae can also enhance oxidation of dissolved iron, simply by producing oxygen as a by-product of their metabolism. Organic material, including algae and plants, is often incorporated into bog iron ores during their accumulation. Some sediment can also be incorporated by stream transport or from local soils.

Bog iron ores are apparently often found underneath bogs at the bottom of breaks in slope. Since they need oxic conditions to develop, it is likely that thick accumulations under peat bogs developed before the peat, but this is a speculation. Bog iron ore can also form on the floors of streams and lakes as encrustations and concretions. Seepages from low-lying poorly-drained ground often produce reddish brown-coloured stains and these would have attracted the attention of early prospectors. Presumably, the more prolific the seepage the more likelihood of finding a nearby concentration of bog iron ore.

The amorphous hydrous ferric precipitates (probably of composition $Fe(OH)_3$ to $FeOOH.nH_2O$), harden on drying out, and progressive dehydration and cementation can take place even in wet conditions. Grain size and crystallinity increase during such 'maturation', leading to the mineral goethite (FeOOH). Ageing over geological time-scales, especially at high ambient temperatures, leads to hematite. This sequence is illustrated using X-ray diffraction analyses (Fig 3). It is unlikely that hematite-bearing bog iron ores would be found in Scotland, due to the recent glaciation.

Bog iron ores are likely to be relatively pure, but with a high content of organic matter. Phosphate, presumably of biochemical origin, may be high; vivianite (iron phosphate hydrate, $Fe_3P_2O_8.H_2O$) is recorded from bog iron ores. Manganese may be associated with bog iron ores but manganese accumulations are likely to be related to locally enriched source rocks such as manganiferous limestones or schists. Reduced aqueous manganese is less readily oxidized than reduced iron, so it is more soluble than iron in conditions of increasing oxygen content. Precipitation of manganese oxyhydroxides therefore requires more extreme oxidizing conditions so it is possible for manganese to travel away in solution while iron is being precipitated. The manganese content of bog iron ore is likely to be very variable depending on the local iron and manganese sources and the precipitation mechanisms. Manganese coatings on, and zoning in bog iron ore (Photos-Jones *et al* 1998), can be explained in general by the fluctuating oxidation state of the fluid. Trace metals which happen to be available in local water may accumulate by absorption onto iron or manganese oxyhydroxides.

Manganese-rich slag is likely to result from the processing of manganiferous bog iron ore, and the manganiferous nodule found at Allt na Ceardaich demonstrates the nature of such manganiferous contamination.

Bog iron ores were divided into two groups by Harder and Johnson (1918): Type 1 are ores formed in marshes, bogs or other shallow depressions, consisting of thin layers of earthy to pisolitic yellow/black iron minerals. They occur on the surface and under several feet of porous soil or peat, and they consist of cemented sand and plant material. Nests of siderite are common. Type 2 consists of ores formed in agitated or flowing waters at lake margins (lake ores). These are usually ooids (2-7mm in diameter) cemented into discs (>20mm in diameter) which coalesce with clastic material to form deposits found at 1-5m depth.

Bog iron ore data was reviewed by James (1966) who mentions the paucity of the modern literature, which comes mainly from Sweden and USSR. Swedish bog iron ore has been discussed by Percy (1886) while, more recently, experimental and analytical work has been carried out on bog iron ores from Wales (Crew and Crew 1995; P Crew pers comm).

Case studies for bog iron ores

The South Erradale, Gairloch , bog iron ore, 'iron dyke' and 'meinn'

On the question of bog iron ore, Macadam (1887) mentions visiting eleven bog iron deposits 'some of them very large', for example at South Erradale, 'where the inhabitants have built a wall, known as the Garadh Iaruim (iron dyke) out of masses.' Analyses of bog iron ore from Gairloch Parish (*ibid*) ranged from 14.58 wt% Fe with 61.08 wt% SiO_2, to good quality ores with 51.57 wt% Fe with 4.72 wt% SiO_2.

Since bog iron ore has been consistently elusive from most sites surveyed or excavated in the Highlands, it was decided to visit and sample the bog iron ore dyke (Fig 2c) referenced by Dixon (1886). The dyke consisted of large lumps of hard rock (about 200-300mm wide) of spongy, almost slag-like appearance, and purple-black in colour. XRD showed that the ore consisted of goethite with moderate crystallinity (Fig 3).

The 'Red Spring' reported by Dixon in the immediate vicinity of the dyke was located and two samples taken. One sample was shown by XRD to consist of moderately crystalline goethite while the other was amorphous (Fig 3). The red clay-like iron-rich material may have been used as a source of ore. It was certainly used for medicinal purposes within recent memory (R Wentworth pers comm).

Early references to extensive bog iron ore in the area, and the reference to gossans (iron caps) in the more recent literature, led us to consider the possible confusion between these two very similar and potentially related types of iron concentration. The trace element chemistry would be expected to be different, and this would have implications for the identification of potential iron ores using slag chemistry. Gossans from stratiform pyritic mineralization could well contain relict sulphur, copper, zinc and possibly lead and gold. Also, while Dixon (1886) and Macadam (1887) dismiss the possible use of local iron ore other than bog iron ore, there does seem a reasonable possibility of local small-scale working of the hematite vein and the magnetite-quartz rock, and this was kept in mind when examining the Fasagh bloomery slags. Potential sources of iron ore for the Fasagh site, Loch Maree are considered further below.

Tamheich Burn, Cowal (NS 030821) with examples of iron seepages
In order to appreciate possible reasons for iron seepages encountered in streams, it is useful to consider the nature of rocks both on the regional and local scales.

The area around Loch Eck and stretching NE and SW along the Cowal peninsula (Fig 1) is within the late Pre-cambrian metamorphic terrain, the Dalradian Supergroup (Johnson 1991) of Scotland. The rocks are mainly regionally metamorphosed metasediments of the Upper Dalradian Southern Highlands Group. The phyllites/schists are dominated by quartz, chlorite and mica. They are not particularly prone to alteration/oxidation to give iron-enrichments, but chlorite weathering does release iron. The Lower and Middle Dalradian rocks further north in the Central Highlands (Fig 1) are by contrast commonly enriched in iron sulphides (Hall 1993). Igneous rocks of various types have intruded the metamorphic terrain. Mafic dykes, typically a few metres across, are common. Such dykes are iron-rich and prone to oxidation to give iron enrichments. There are also faults and fracture zones, and alteration and iron enrichments could be localized in such areas. However, glaciation means that surface and deep alteration was removed.

Dalradian chloritic phyllites/schists are well exposed in the floor and banks of the Tamheich Burn. The surrounding alluvial sediment must only be a metre or so thick. There are many small iron seepages coming from the banks of the stream. Some of the seepages come from fractures in the schists but some are draining the schist/alluvial sediment contact. Local fracturing and weathering of the chlorite schist, the raised nature of the bedrock/alluvial sediment contact and local acidic ground-water could be reasons for the enhanced iron mobility.

Extensive but superficial iron staining in the streams could have attracted the iron smelters and prospectors to examine the area more closely. Crusts of iron oxyhydroxides a few millimetres thick are sufficient to cement stream gravel. The deposits appear to be too small to justify exploitation, but larger accumulations of iron (bog-iron ore) representing thousands of years of seepage, forming perhaps a few cubic metres, could have been removed by the iron smelters and remained unreplenished. Small areas of the stream bank close to the bloomery mounds appear to have been cut back, possibly in an effort to work concentrations of iron which were perhaps suspected to continue under the thin soil cover.

Ferric oxyhydroxide replacement encrustations

Replacement and encrustation of other minerals by ferric oxyhydroxides and hematite in a laterization-like process may lead to iron ore deposits, but these are likely to be small in Scotland because of glacial erosion and the cool climate. The chemical processes of dissolution and precipitation by oxidation are very similar to that outlined for bog iron ores, but there is no significant transport of the iron.

Ferrous-iron-rich minerals such as pyrite (FeS_2) and siderite ($FeCO_3$) are very prone to replacement by ferric oxyhydroxides. Rocks and veins which are particularly rich in these minerals can be altered on deep weathering to produce viable iron ores.

Calcite ($CaCO_3$) is also prone to replacement by ferric oxyhydroxides because of neutralization by calcite of iron-rich acidic ground-water. Calcite veins and calcite minerals may therefore be replaced. This can lead to very small accumulations of iron which may attract a prospector, but which would prove to be very superficial or localized.

Case study: Strone Road End, Cowal (NR 943835)
The bloomery site consists of a poorly exposed mound and building foundations on hill slopes adjacent to the deep gorge of Kilail Burn. Evidence of iron encrustations were located in the gorge.

The area lies within the middle Dalradian Argyll Group. The metamorphic rocks tend to be mainly chloritic schists, but containing calcareous material. The metamorphic rocks of the Argyll group are more diverse in composition than those of the Southern Highlands group and are often enriched in pyrite, due to the nature of the depositional environment and localised synsedimentary mineralization (Hall 1993). There are various later minor igneous intrusions, and faulting and fracturing.

The local metamorphic lithologies are well exposed in the stream gorge running NE from the site. A reconnaissance investigation revealed large boulders of iron-stained carbonate-cemented breccias in the stream. There is therefore the possibility of a local source of iron ore. The Geological Survey memoir reports iron carbonate veining in the Cowal area (Hill 1905). Reddish-brown calcite is easily mis-identified in the field as iron carbonate, and the mineral can be simply iron-stained calcite, or calcite extensively replaced by iron oxyhydroxides. However, it is not impossible that such local iron enrichments could have reached ore grade for bloomery-scale iron production. A sample of carbonate analysed by XRD proved to be calcite with only very minor hematite. Although iron-enriched, the material would be unsuitable (too low grade) as an iron ore.

Gossans

Major accumulations of ferric oxyhydroxides produced by replacement of iron-rich source rocks are known as gossans or iron caps, and have long been known to occur above many types of ore deposit. This is because iron sulphides often accompany other metal sulphides such as galena (PbS) and sphalerite (ZnS). Gossanous iron ores usually contain evidence of their original nature. This may be in the form of textures inherited from the original rock. Also, minerals such as galena may remain as insoluble relics, and secondary minerals such as cerrusite ($PbCO_3$), which reflect the chemistry of the original rock, may form. Gold is often concentrated from the source rock into the gossan, from which it can be panned. Early prospectors could well have had such associated minerals in mind when they were seeking iron ore deposits.

Gossans (strictly *in-situ* accumulations of iron) and bog iron ore (precipitated from solution) form a spectrum of related and very similar types of iron ore. In some cases it is difficult to tell how much iron transport has been involved (Nicholson undated).

None of the bloomery sites being investigated were located close to gossans, although gossanous developments are well known in the Gairloch area, south west of the Fasagh site at Loch Maree (see below).

Hematite ores

Hematite (Fe_2O_3) is quite common in Scotland as a component of veins which can cut any type of rock. It can form up to 100% of fracture infills, especially in small fractures. The hematite can occur as black metallic coarse crystalline plates or fine crystalline aggregates. Reddish-brown coated mamillary/botryoidal forms of fine metallic hematite (like compressed bunches of grapes) are common. Fine grained reddish-brown hematite is often rich in fine silica. Quartz veins in metamorphic rocks may contain pockets rich in hematite. Some rocks can be hematized; that is, they contain hematite in fractures and fine hematite impregnating the rock. Some unusual metamorphosed sedimentary rocks may be rich in hematite. Hematite veins at Muirkirk in the Southern Uplands (Fig 2b) and the Lecht (with manganese, Nicholson undated) in the Grampian Highlands were worked in the 1730s. The Muirkirk ore was smelted nearby at Tarrioch Furnace, close to Nether Wellwood, and later transported to Bonawe, while the Lecht ore was smelted at Culnakyle near Abernethy (MacGregor *et al* 1920), the site of an early charcoal blast furnace (*c*1756).

Case study:Allt na Caerdaich, Cowal (NS 12459275)

The setting is as for Tamheich Burn above. There is a hematite vein at Strachur, about 8 km NNW of Allt na Ceardaich, which could have provided some local iron ore for the bloomery workings at Allt na Ceardaich. Macadam (1881) reports the analyses of two ore specimens, picked as pure as possible, and one specimen found in the veins at Strachur. The pure samples contained 88.83 wt% Fe_2O_3 with silica as the main impurity, while the vein material only contained 37.52 wt% Fe_2O_3. Mineralization in the area is not well researched or documented. There could therefore be small hematite veins in the vicinity of Allt na Ceardaich.

The metamorphic rocks are well exposed in Allt na Ceardaich from close to the shore of Loch Eck upstream for about 400m. Some slag was noted on the shore where the stream enters Loch Eck. The metamorphic rocks are siliceous chloritic phyllites which outcrop as gently dipping slabs. Thin quartz veins and pods, typical of such phyllites, are found both in outcrop in the stream and as pebbles and boulders. Black hematite with a typical reddish tinge, in quartz veins, is probably primary, and is a pointer to the possible existence of hematite enrichments in the area. Rather powdery yellow/brown limonite in quartz probably represents oxidized pyrite and/or iron-replacement of calcite. Calcite was found in a few veins, but pyrite was not confirmed. Slabs of phyllite have possibly been quarried on a small scale at an outcrop immediately upstream from the second bloomery site. On a shelf cut into the slopes above the second bloomery, and adjacent to the stream, there is a quarried outcrop of an igneous rock, probably a diorite sheet (2m thick), underlying chlorite schists. It seems most probable that the diorite was quarried as a local building stone or road metal. The soil is locally very reddish brown, but no potential bog iron ore was detected in stream banks or forestry road cuttings. There is potential for small bog iron ore concentrations in the undulating area in the hills above the bloomeries, and although hematite remains a possible ore, the manganiferous nature of the bloomery slags suggests bog ore was used at this locality.

Clayband and blackband ironstones

These are sedimentary ironstones of diagenetic/bacteriogenic origin formed of impure fine-grained siderite ($FeCO_3$). They are common in rocks of Carboniferous age in Scotland, especially in coal measures. The clayband ironstones are concretions which can be up to 1m across, found in shales. The blackband ironstones are continuous layers. The siderite can contain sedimentary mineral impurities, *eg* quartz, kaolin, illite clay and sometimes pyrite. The blackband ores are more difficult to smelt. Sideritic ironstones occur throughout the Midland Valley of Scotland and clayband ores were smelted using coal from 1760, while blackband ores were not used until the 19th century. But clayband ores from Fife were reportedly used at Loch Maree in the first half of the 17th century (MacGregor *et al* 1920). Although the ideal composition of siderite is $FeCO_{3,}$ it can contain some magnesium and/or manganese, as well as the possibility of fossils, and this provides some potential for fingerprinting siderite sources.

Macadam (1887) saw only fragments of bog iron ore at Fasagh, but found both hematite of Cumberland type (probably from NW England) and Carboniferous clayband ironstone (siderite, probably from the Coal Measures of Midland Valley of Scotland) at both Letterewe and Red Smiddy. He concluded that the remains of stockpiles of iron ore provided evidence that the ore was shipped to Poolewe *en route* to Letterewe.

Metasediments: magnetite schists (Banded Iron Formation) and pyritic schists

Many metamorphic rocks contain minor amounts of magnetite but unusual enrichments occur in the magnetite schists of the Gairloch/Loch Maree area, NW Scotland (Fig 1). The banded schistose rocks contain magnetite with fine quartz and various iron silicates, mainly amphiboles. Because of their banded nature they have been called banded iron formation (BIF), but they are not similar in origin to the major Precambrian Iron Formations (Superior Type BIF) of the world (James 1992). These are thick sequences of finely bedded quartz hematite beds which extend for kilometres. Not only are they major iron ore deposits, but their occurrence has important implications about the evolution of the oxygen content of the atmosphere. There is a category of BIF known as the Algoma Type and the magnetite schists of NW Scotland probably belong to this group. The Algoma Type is more limited in scale and is sometimes associated with volcanics and beds of massive sulphide, including copper, mineralization. Pyritic schists are in fact also found in the Gairloch/Loch Maree area and, as well as potential sources of pyrite, they could represent a primary iron-source for local gossans and bog iron ores.

Case study: Loch Maree and the Fasagh site (NH 011654)
The presence of the Loch Maree 'furnaces' of early 17th century date led to the suggestion that there could be a local source of iron ore. However, the literature is contradictory as to whether local ore was mined or not. It was said that 'the only deposits known consist of small pans of bog iron ore. Any pans of local bog iron are assumed now to have been worked out' (MacGregor *et al* 1920). Nevertheless, Macadam (1887) and Dixon (1886) perhaps had the best knowledge of local iron ores, and their comments are as a result the more helpful.

Macadam mentions that there are recorded traditions that local iron ores other than bog iron were employed in the area, but he was suspicious of such reports because of the frequent confusion of the term 'works' and 'mines'. Also, he visited outcrops on the side of Slioch (Fig 5) which were possible iron workings, and found only low-grade pyritic deposits with no more than about 8 wt% Fe. The outcrop (Dixon 1886) is a 'large band of ferruginous stone that runs from Letterewe, in a southeasterly direction, along the shores of Loch Maree to the further end of the base of Slioch. It is so extensive and so rusty in colour, that it can be easily discerned from the county road on the opposite side of the loch.' It also occurs in other parts of the Gairloch Parish where local people report it was quarried, *viz:* on the south side of the Furnace burn at Letterewe, nearly a quarter of a mile above the site of the iron furnace; on the face of the ridge immediately behind and above the cultivated land at Innis Ghlas; at Coppachy; and in a gully, called Clais na Leac, at the NW end of the cultivated land at Smiorsair (*ibid*). Dixon however considers that there is no sign of working at these localities where there are only natural outcrops of ferruginous rock and concludes that 'we may therefore dismiss the tradition that iron ore was obtained directly from these supposed quarries as not only unreliable but impossible.'

This brief review is mainly based on Park (1978 and 1991). Loch Maree straddles an extensive complex belt of Precambrian rocks in NW Scotland known as the Lewisian after the type area, Isle of Lewis. While the dominant rocks of the Lewisian are high-grade grey gneisses of graniodioritic composition which could well represent metamorphosed igneous rock, there are also schistose metasediments forming two belts, one stretching SE from Gairloch and one on the N side of Loch Maree. These belts constitute the Loch Maree Group which is believed to be of Lower Proterozoic age, about 2.4 thousand million years old. The group covers an area of about $130km^2$, is up to 5km thick and consists of folded semipelitic quartz-plagioclase mica schists with thin layers of marble, graphite schist and some 'unusual' metasediments (magnetite schists and pyritic schists) as well as thick sequences of amphibolite which represent metamorphic mafic igneous material of intrusive and/or

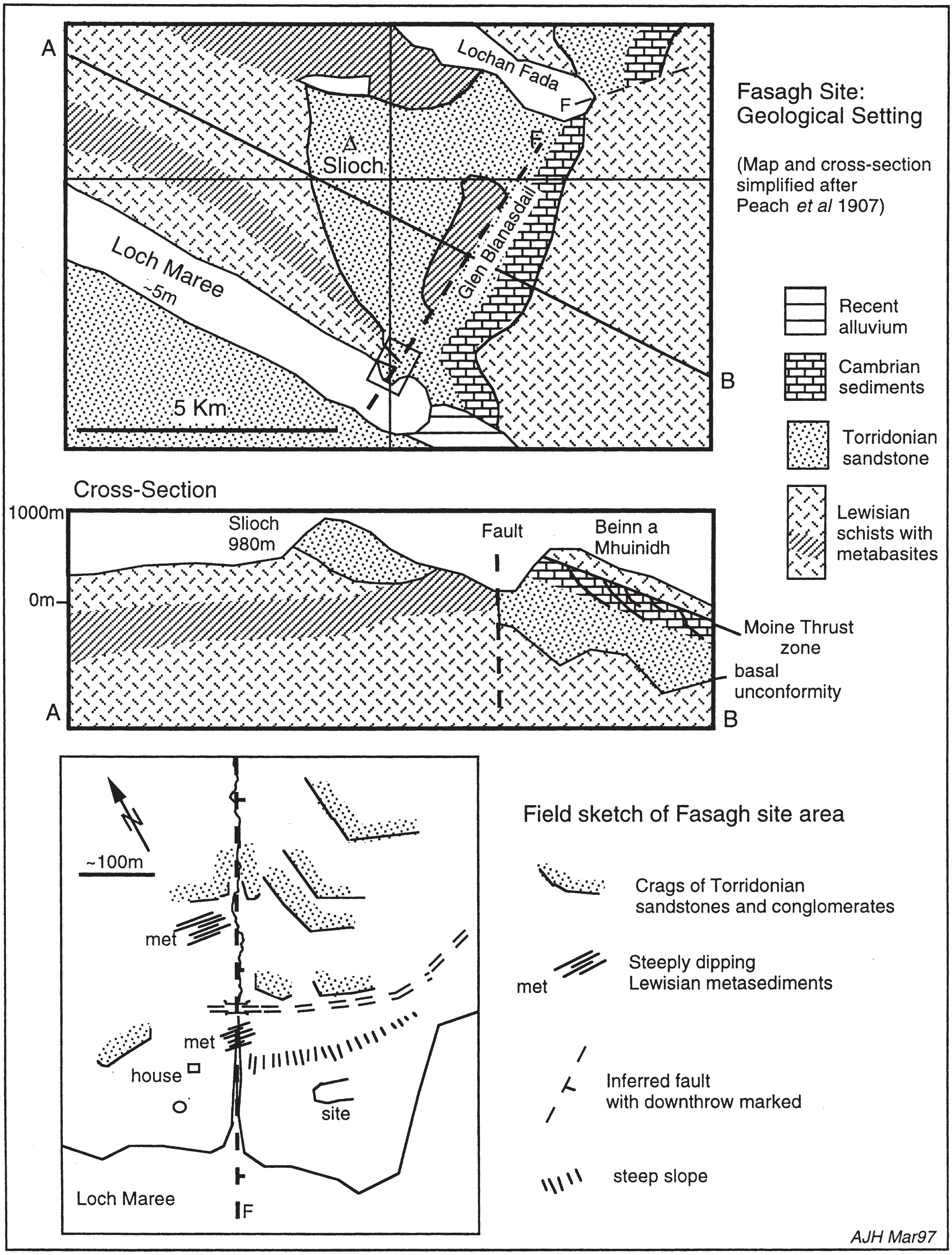

Figure 5: Geological setting of the Fasagh Site, Loch Maree and field sketch of the Fasagh site area showing main rock outcrops. The geological map and cross-section are simplified after Peach et al *1907.*

volcanic origin.

The Lewisian is overlain unconformably by a late Precambrian thick coarse feldspathic sandstone sequence known as the Torridonian and this is present at the eastern end of Loch Maree (Fig 5). It is the 'unusual' rocks of the Loch Maree metasedimentary group which offer the possibility of local iron ores, of at least four different types. These are best known from the more accessible area SE of Gairloch, but MacGregor *et al* (1920) mention the occurrence of the BIF and pyritic graphitic schists on the north side of Loch Maree:

i) The BIF consists of discontinuous thin beds to pods of folded rock made of alternating layers of coarse quartz and magnetite (Fig 2a). A representative analysis of this rock as exposed at Kerrysdale, Gairloch, gave, in wt%: SiO_2 53.77; TiO_2 0.07; Al_2O_3 0.24; Fe_2O_3 22.77; FeO 16.89; MnO 0.84; CaO 0.94; MgO 0.75; Na_2O 0.07; P_2O_5 0.63; K_2O, F, S and Cl not detected (Al-Ameen 1979 in Park 1991). Crushing of this hard but brittle rock would have been possible, to provide nearly pure magnetite feedstock. Some siliceous BIF is evidently manganiferous with 6.60 wt% Mn reported (*ibid*).

This magnetite-rich rock has several distinctive textural and mineralogical features which would make it easy to identify if found associated with an iron-smelting site. A representative sample of banded magnetite-rich rock from near Gairloch proved to contain coarse-grained magnetite (Fig 2a) with very minor pyrite, associated with fine-grained cherty quartz and iron-rich amphiboles. This magnetite rock was a potential constituent of the rusty and magnetic building material of the anvil at Fasagh but this proved to be dominated by magnetic wustite (Photos-Jones *et al* 1998) with no evidence for use of natural magnetite.

ii) Pyritic quartz-carbonate rocks (3-4m thick) occur about 6km SE of Gairloch. Because of their enhanced copper, zinc, lead and gold contents, these rocks have been assessed as potential ore deposits (Jones *et al* 1983 and 1987). The mineralization is believed to be of stratiform exhalative volcanic-associated copper-zinc type which is often found associated with high level mafic magmatism in volcano-sedimentary environments. Iron sulphide in the form of pyrite and/or pyrrhotite may be enriched enough in places to have been exploited as an iron ore.

iii) The sulphide-rich carbonates and the pyritic graphitic schists of the Loch Maree metasediments could provide an exceptional source of local secondary iron, and in places there is evidence of the development of gossans. These are essentially concentrations of ferric oxyhydroxides formed on top of the weathering iron-rich rock and which usually contain relict material indicative of the nature of the primary source. Jones *et al* (1983) report the presence of such gossanous sulphide-rich horizons in the area and proved details of a 100mm wide gossan developed within a sequence of mafic schists on Teangadh Bhuidhe Mhor; the gossan contains copper, zinc and trace gold. However, gossanous layers up to 40m across and extended over 6km developed over stratiform iron sulphides at Sidhean Mor, about 5km SE of Gairloch, are also mentioned by Jones *et al* (1987).

iv) The iron-sulphide-rich metasediments, as well as rocks such as graphitic schists with low iron sulphide concentrations, provide a local bedrock of generally enhanced iron content. On weathering, dissolution and transport, this iron becomes available for bog iron ore development.

The Fasagh site is situated on a small alluvial promontory at the mouth of the Fasagh river where it enters Loch Maree (Fig 5). The surrounding area has been mapped by the British Geological Survey (Peach *et al* 1907). Figure 5 is simplified after this geological map and cross section. The regional basement consists of Precambrian Lewisian schists/gneisses, and the Loch Maree metasediments occur along the north side of Loch Maree. The Lewisian basement lithologies are overlain unconformably by sandstones and by some basal conglomerates of Precambrian Torridonian age. The continental, generally red/brown oxidized feldspathic sediments of the Torridonian, infilled and buried a Precambrian landscape developed on the Lewisian basement. This partially exhumed fossil topography is evident in the cross-section of Figure 5. The Torridonian is in turn overlain unconformably by a sequence of Cambrian sandstones, shales and dolomitic limestones. These are overlain by the much older Lewisian which has been thrust over the Lewisian/Torridonian/Cambrian sequence from the east. The geology is further complicated by faulting.

A search for iron ores was made around the shoreline to the south of the Fasagh site. Most of the rock types of the surrounding area were noted on the pebble beach, but no iron ore was found. Possible iron ores from this area would have been magnetite schist, schistose pyrite, vein hematite, gossanous rock after pyrite and bog iron ore. Also, a pisolitic iron ore developed in Cambrian dolomitic limestone/shale rocks was reported to occur on the northern slopes of Meall a'Ghubhais, WNW of Kinlochewe and elsewhere in the region (Peach *et al* 1907). However this is only 100-150mm thick. Imported ore might have been mamillary vein hematite from Cumberland or sideritic ore from Fife. No iron ores were noted in the rocks encountered in the trial trenching at Fasagh.

A reconnaissance examination of the rocks exposed to the N of the site (Fig 5) confirmed the presence of Lewisian metasediments. These are dark laminated cherty rocks with no pyrite evident. The Torridonian rocks are coarse sandstones in thick beds which result in a stepped crag outcrop on the hillside. These dip about 10-15° to the NE. The coarse sandstones close to the road and bridge are rather pale compared with normal Torridonian and may be leached of their iron. Both the basement Lewisian (metasediments and metabasites) and the Torridonian sandstones are potential sources for the production of bog iron ore on weathering. It seems quite likely that bog iron ore could be developed in the surrounding area, *eg* in Glen Blanasdail.

The details of the local rock types are also potentially relevant to the archaeological investigation of the nature of the unusual ferruginous building material used for the anvils at Fasagh.

Conclusion

The early prospectors for iron ore deposits for bloomeries in the Scottish Highlands could have developed an observational expertise which would have helped them to locate different styles of mineralization. There were several types or iron ore available, and a spectrum of bog iron type ores which are likely to be of quite variable nature and size. It is the regional and local geological setting of a bloomery site which dictates what types of iron ore deposits could have been available, and this knowledge can guide the archaeologist in the search for evidence for the nature of iron ore used at bloomery sites.

In practice, it has not been easy to establish for certain what sources of iron were used for the bloomery sites under investigation. The small scale of operations means that there is little chance of finding the remains of stockpiles of ore, and small-scale local working of iron ore deposits, particularly bog iron ore, would leave little trace. Nevertheless, this geological study has demonstrated that there is potential for iron ores other than bog iron ores, and use of bog iron should not just be assumed without confirmation. The various potential iron ores do have distinctive mineralogical and petrographic characteristics and a careful search of bloomery sites for fragments of ore is a valid approach. Bog iron ores are particularly friable and in the absence of any positive petrographic evidence of ore-type, but with chemical evidence from the slag for use of manganiferous ore with minor silica as the main impurity (Photos-Jones *et al* 1998, 23-6), it seems reasonable to conclude that bog iron ore was the only ore used. This seems to have been the case at these bloomery sites.

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