



# Petrology and geochemistry of the ~2.9 Ga Itilliarsuk banded iron formation and associated supracrustal rocks, West Greenland: Source characteristics and depositional environment

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## ARTICLE INFO

### Article history:

Received 22 May 2011

Received in revised form 12 March 2012

Accepted 18 April 2012

Available online 5 May 2012

### Keywords:

West Greenland

Archaean

Itilliarsuk BIF

Supracrustals

Seawater chemistry

Neodymium isotopes

## ABSTRACT

Here we present new field, petrographic and geochemical data from the ~2.9 Ga Itilliarsuk banded iron formation (BIF) and associated lithologies within the Itilliarsuk supracrustal belt, south-eastern Nussuaq, West Greenland. The supracrustals represent a volcanic–sedimentary sequence, which rests unconformably on a basement of tonalite–trondhjemite–granodiorite (TTG) lithologies. Felsic metagreywackes, meta-semipelites and thinly bedded ferruginous shales were identified intercalated with the Itilliarsuk BIF. Other associated rocks include metapelites, acidic metavolcanics and metagabbroic sills. The supracrustals have experienced amphibolite-facies metamorphism, which has resulted in complete resetting of the U–Pb system with an apparent age of  $1895 \pm 48$  Ma. This tectono-metamorphic event corresponds well with the Paleoproterozoic Rinkian orogeny known from this region. The Itilliarsuk-(oxide-facies) BIF has been divided into two segments on the basis of major and trace elements chemistry: a shaley-BIF with a strong clastic component and a more chemically pure BIF. The shaley-BIF contains high terrigenous influx as reflected by elevated  $\text{Al}_2\text{O}_3$  (up to 12 wt.%),  $\text{TiO}_2$ , high field strength elements (HFSE) and transition metals. The chemically pure BIF is characterised by alternating high iron (~68 wt.%) and high silica (~64 wt.%) bands with low total rare earths and yttrium (REY),  $\text{Al}_2\text{O}_3$ ,  $\text{TiO}_2$  and HFSE contents, suggesting a low detrital component. The least altered bands of the BIF record diagnostic Archaean seawater features with Post-Archaean Average Shale (PAAS)-normalised positive La- and Eu-anomalies, enrichment in heavy rare earth elements (HREE) relative to light rare earth elements (LREE) [ $(\text{Pr}/\text{Yb})_{\text{PAAS}} < 1$ ], and absence of Ce-anomalies which suggests deposition from an anoxic water column. Sm–Nd isotopes of the chemically pure silica-rich bands record  $T_{\text{DM}}$  model ages of 3.23–2.85 Ga and initial  $\varepsilon_{\text{Nd}}$  ( $\varepsilon_{\text{Nd}}(i)$ ) values in the range of +0.43 to +4.05, average of +1.35. In contrast, the chemically pure Fe-rich bands yield  $T_{\text{DM}}$  model ages of 3.61–3.22 Ga and  $\varepsilon_{\text{Nd}}(i)$  values from –2.87 to +0.09, average of –1.29. The associated supracrustal rocks in the study area have significantly higher, positive  $\varepsilon_{\text{Nd}}(i)$  values. The  $^{143}\text{Nd}/^{144}\text{Nd}$  in the Itilliarsuk BIF, therefore, contrasts world BIFs by exhibiting radiogenic, positive  $\varepsilon_{\text{Nd}}(i)$  values in shallow seawaters where the REY's were controlled by a local, depleted continental crust, whereas the negative  $\varepsilon_{\text{Nd}}(i)$  values found in the iron-rich layers suggest that the submarine hydrothermal source was influenced by an enriched mantle, possibly an older subcontinental lithospheric segment. The felsic metagreywackes are immature, first-cycle ( $\text{SiO}_2/\text{Al}_2\text{O}_3 \sim 4.4$ ,  $[\text{La}/\text{Yb}]_{\text{CHON}} > 1$ ) metasediments with affinities to TTG-suites, primarily extrusives, whereas the meta-semipelites and metapelites contain a larger mafic contribution with higher content of  $\text{Fe}_2\text{O}_3$ , MgO, Cr, Ni and HREEs. This suggests that the BIF was deposited in a highly unstable basin, presumably in a palaeo-continental slope or outer shelf environment, with frequent fluctuations of epiclastic and volcanogenic sediments derived from adjacent bimodal sources. The  $T_{\text{DM}}$  model ages and the use of Th–Sc–Zr and La–Th–Sc tectonic discrimination plots indicate that the metasediments were sourced from a juvenile ocean island arc setting.

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## 1. Introduction

Banded iron formations (BIFs) are chemical sediments that precipitated throughout the Archaean and early Paleoproterozoic. The majority of BIFs consist of alternating silica-rich and iron-rich bands that range from small-scale laminated microbands (few mm), through mesobands ( $\geq 10$  mm) and up to metre-scale macrobands (Trendall and Blockley, 2004). The mineral composition of BIF has been modified by diagenetic and metamorphic overprinting, and therefore, the main mineral phases now found in BIF, such as hematite ( $\text{Fe}_2\text{O}_3$ ), magnetite ( $\text{Fe}_3\text{O}_4$ ), microcrystalline quartz ( $\text{SiO}_2$ ), stilpnomelane ( $\text{K}(\text{Fe,Mg})_8(\text{Si,Al})_{12}(\text{O,OH})_{27}$ ), Fe-amphiboles, calcite and dolomite-ankerite are actually of secondary origin. Proposed primary minerals are ferric hydroxide ( $\text{Fe}(\text{OH})_3$ ), siderite ( $\text{FeCO}_3$ ), greenalite ( $(\text{Fe}_3\text{Si}_2\text{O}_5(\text{OH})_4$ ) and amorphous silica (Klein, 2005). The bulk of the iron in BIF originated as dissolved Fe(II) from submarine hydrothermal vents and was subsequently transformed to dissolve Fe(III) in the upper water column by either abiological or biological oxidation. The ferric iron then hydrolysed rapidly to ferric hydroxide and settled to the sea floor where further transformations ensued (Bekker et al., 2010). Silica was either sourced from hydrothermal venting (Steinhöfel et al., 2010) or from continental weathering (Hamade et al., 2003).

As marine chemical sediments, the trace element and isotopic composition of BIF have been used as proxies for understanding Earth's ancient surface environment. In particular, several recent studies have used BIF to constrain the preceding period of arguably the most important transition in Earth's history, that being the 'Great Oxidation Event' (GOE) at around 2.5 billion years ago when the atmosphere became oxygenated. For instance, temporal trends in BIF Ni content have led to the suggestion that the oceans before 2.7 Ga could have supported the growth of methane-producing bacteria throughout much of the water column, but a significant decline in seawater Ni concentrations thereafter led to a methanogen Ni-famine, a subsequent drop in methane production, and ultimately a rise in atmospheric  $\text{O}_2$  as oxygen-producing cyanobacteria expanded deeper into the water column left vacant by the methanogens (Konhauser et al., 2009). Based on Cr isotope values in BIF through time, Frei et al. (2009) found that cyanobacterial activity may already have been abundant by 2.8 Ga ago, and that their production of  $\text{O}_2$  may have led to Earth's first oxidative weathering of the continents several hundred millions years earlier than the GOE. Supporting evidence for oxygenated marine surface waters between 2.8 and 2.7 Ga comes from  $^{13}\text{C}$  enrichment in kerogens (Eigenbrode and Freeman, 2006), cyanobacterial and methanotrophic biomarkers in shales and carbonates (Eigenbrode et al., 2008), high Re concentrations in black shales (Kendall et al., 2010) and N isotopic values in kerogens indicative of a coupled nitrification–denitrification cycle (Godfrey and Falkowski, 2009). However, relative to the GOE, the oxidative weathering during the Mesoarchaeon may not have been very profound as suggested by generally lower authigenic Cr supply to the oceans (Konhauser et al., 2011).

As the timing for the spread of cyanobacteria, and the oxidation of the surface environment, is being pushed back in time, it becomes critical to analyse BIF from strata older than 2.8 Ga to assess the composition of seawater and the impact of continental weathering vs. submarine hydrothermal fluids on the source of solutes, and ultimately the diversity of the marine biosphere at that time. Previous work attempting to ascertain the composition of Mesoarchaeon (3.2–2.8 Ga) oceans has focused on rare earth element and yttrium (REY) patterns of individual bands within BIF. With few exceptions, Post-Archaean Average Shale (PAAS)-normalised REY patterns of BIF are similar to those of modern seawater, exhibiting low REY contents, positive La- and Y-anomalies and depletion of light rare earth elements (LREEs) relative to heavy rare earth elements

(HREEs) (Bau and Dulski, 1996; Bolhar et al., 2004; Johannesson et al., 2006). The exceptions are that Archaean BIFs have positive Eu-anomalies and they do not show the characteristic strongly negative Ce-anomalies of modern seawater. The former is attributed to high-temperature ( $>250^\circ\text{C}$ ) hydrothermal fluids from mid-ocean ridges and back-arc spreading zones injecting solutes into anoxic bottom waters (Bau and Möller, 1993), while the latter reflects the lack of significant oxidizing agents (e.g.  $\text{O}_2$ ) to fractionate Ce(III) from neighbouring REEs (Bolhar et al., 2004; Rollinson, 2007). Neodymium (Nd) isotopes have also shown to be highly valuable in tracing the continental vs. hydrothermal input to BIF. For example, the 2.9 Ga Nemo BIF in North America records mixing of  $^{143}\text{Nd}/^{144}\text{Nd}$  from primarily two sources: (1) anoxic deep water with generally positive initial  $\varepsilon_{\text{Nd}}$  ( $\varepsilon_{\text{Nd}}(\text{i})$ ) values, reflecting submarine MORB-like depleted sources, and (2) shallower waters with generally lower  $\varepsilon_{\text{Nd}}(\text{i})$ , reflecting riverine input from evolved continental sources (Frei et al., 2008). Newly compiled  $^{143}\text{Nd}/^{144}\text{Nd}$  data by Alexander et al. (2009) of BIF older than 2.7 Ga suggests that bulk anoxic Archaean seawater was dominated by high-temperature hydrothermal alteration of mantle-derived oceanic crust, presumably also responsible for delivering the iron to the BIF. However, deviation from this mantle Nd signal has been recorded in the 2.9 Ga Pongola BIF in South Africa which was deposited in a near-shore, shallow-water environment where continental fluxes of Fe and other solutes contributed greatly to the unit's composition (Alexander et al., 2009).

Despite the previous studies on Mesoarchaeon BIF that have focused on the source of the BIF components, there is a paucity of information pertaining to the trace element composition of those BIF, and in particular, what they might tell us about ancient seawater chemistry. In this regard, we have investigated the composition of the relatively unexplored Itilliarsuk BIF exposed within the Mesoarchaeon Itilliarsuk supracrustal belt on south-eastern Nuussuaq, West Greenland. Major and trace elements whole-rock geochemical analyses, as well as Nd and Pb isotope analyses have been carried out on individual micro- and mesobands from the BIF and associated lithologies. In addition to the BIF, the interbedded clastic metasediments have also been analysed in order to draw an interpretation of the depositional environment of the Itilliarsuk BIF. We show here that the BIF was deposited in an anoxic water column, with no evidence for surface water oxygenation, and by extension, abundant cyanobacterial activity.

## 2. Geological background

The Itilliarsuk BIF is exposed in the promontory called the Itilliarsuup Qaqqaa (Fig. 1). The area comprises the western part of a 25 km, ENE–WSW trending belt of Archaean supracrustals called the Itilliarsuk supracrustal rocks. The belt belongs to the southern part of the Nuussuaq domain and is located along the northern shore of the Torsukattak ice-fjord, on the south-east Nuussuaq (Fig. 1). With a total of 2.5 km, the Itilliarsuup Qaqqaa is the thickest sequence in the Itilliarsuk supracrustals (Garde and Steenfelt, 1999). Presented in Fig. 2A is a field photo with an overview of Itilliarsuup Qaqqaa and the main lithologies; the main lithologies are placed into the general stratigraphical column in Fig. 2B.

The main structural deformation and the metamorphic overprinting of the Nuussuaq domain developed primarily during the early Proterozoic Rinkian orogenic event, ca. 1.7–1.8 Ga ago (Garde and Steenfelt, 1999). A SSE-dipping Palaeoproterozoic shear zone along the Torsukattak ice-fjord divides the Nuussuaq domain from the southern Ataa domain. Evidence from the acidic metavolcanic unit at the southern part of Itilliarsuup Qaqqaa shows down throw of a southern hanging wall towards the south-east. This normal fault developed during the Rinkian orogeny, and accounts for

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