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# Pb isotopes during crustal melting and magma mingling — A cautionary tale from the Miki Fjord macrodike, central east Greenland

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#### ABSTRACT

Pb isotopic data are presented for hybrid rocks formed by mingling between mantle-derived tholeiitic magma of the Eocene Miki Fjord macrodike (East Greenland) and melt derived from the adjacent Precambrian basement. Bulk mixing and AFC processes between end-members readily identified in the field fail to model the Pb isotope systematics. Selective contamination during diffusional exchange, which can explain the complex Sr and Nd isotope compositions of the hybrid rocks (Blichert-Toft et al., 1992), cannot fully account for the variability of the Pb isotopic data using the identified crustal end-members. The crustal anatectic end-member, although similar in Sr and Nd isotope composition, has a markedly different Pb isotopic composition than its source gneiss. The differences are consistent with preferential incorporation of radiogenic Pb from accessory phases such as metamict zircon or loosely-bound Pb from grain boundaries during disequilibrium melting of the host rock by the mafic magma. The crustal melt involved in magma interactions was therefore heterogeneous with respect to Pb isotopes on a metre-scale. These results illustrate the difficulties inherent in interpreting isotopic variations in contaminated mafic magmas even when the end-members are well constrained by field relations. We show that the Pb isotopic composition of the crustal contaminants and contamination trajectories for the Miki Fjord hybrid magmatic lithologies are markedly different from regional basement gneisses and contaminated lavas of the associated flood basalt province that were contaminated by characteristically unradiogenic Pb and Sr lithologies resembling Lewisian basement not exposed locally.

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#### 1. Introduction

Crustal contamination is a fundamental process affecting the composition and evolution of mantle-derived magmas emplaced in continental settings, and is most easily identified through variations in the radiogenic isotopic compositions of igneous rocks and/or their mineral constituents. The often large differences in isotopic composition and elemental concentrations between crust and magma result in large variations in isotopic compositions of contaminated magmas with only small degrees of crustal interaction. However, in many cases, the exact nature of the contaminant is unknown, and the processes by which contamination occurs (bulk-assimilation, assimilation of partial melts of crust and/or diffusive exchange) are often poorly constrained. Pb isotopes have provided overwhelming evidence for crustal contamination for at least some of the voluminous

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flood basalts erupted in central East Greenland during the opening of the North Atlantic (e.g. Thirlwall et al., 1994; Fitton et al., 1998; Saunders et al., 1999: Hansen and Nielsen, 1999: Ellam and Stuart, 2000: Andreasen et al., 2004: Peate et al., 2008). In this study we examine the Pb isotopic systematics of the Miki Fjord macrodike in central East Greenland where interactions between a basaltic magma mingled with a partial melt of the adjacent crustal component have been preserved in situ and end-members together with intermediate hybrids are clearly definable in the field. The locality represents a classic field example of contamination of mafic magmas by partial melts derived from the surrounding country rocks to which mathematical models such as bulk-mixing and/or combined assimilation and fractional crystallisation (AFC) are typically applied. The present study was carried out for several purposes: firstly as a general investigation of how the Pb isotopic system behaves during mingling and contamination between two identifiable chemically and rheologically distinct magmas; secondly because the Miki Fjord macrodike is often cited as a type-example of selective diffusive exchange between co-existing melts (Rosing et al., 1989; Blichert-Toft et al., 1992); and thirdly because the Miki Fjord macrodike and other similar bodies in the vicinity (e.g. Geist and White, 1994; Momme and

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Wilson, 2002) represent potential feeder systems and/or magmas contemporaneous with, and potentially related to, the intensely-studied Skærgaard intrusion and voluminous flood basalt sequences. We therefore also evaluate our results in the light of contamination trends for magmas emplaced elsewhere in central East Greenland.

The target of this investigation is the Soedalan segment of the Miki Fjord macrodike (Blichert-Toft et al., 1992) where hybrids formed by interactions and mingling between mantle-derived tholeiitic magmas and locally-derived crustal melts have been frozen in situ. The large age difference between the macrodike, emplaced during Eocene flood volcanism at ca. 55 Ma, and the Precambrian (~2.9 Ga) gneiss hosting the intrusion, has led to marked contrasts in isotopic composition between the intrusion and host rock that facilitate tracking contamination processes. This study is an extension of the investigations of Blichert-Toft et al. (1992) who found that co-variations of Sr and Nd isotopic compositions in the hybrid magmas are not adequately described by bulk mixing or AFC models. They instead demonstrated that decoupling of these two isotopic systems due to differences in the self diffusivities for Sr and Nd, as observed experimentally (Lesher 1990, 1994), and consequent selective exchange between mingling mafic and felsic magmas, was an important process. Several other studies have also attributed apparent decoupling of Sr and Nd isotope systematics to differences in the diffusion rates (e.g. Halliday et al., 1984; Wörner et al., 1985; Holden et al., 1987, 1991; Pin et al., 1990; Allen, 1991; Waight et al., 2001), however, to our knowledge the behaviour of Pb in such systems has not yet been examined. Examining the Pb isotopic systematics of these same samples will provide further constraints on the selective contamination hypothesis of Blichert-Toft et al. (1992), and ascertain whether processes other than, or in addition to, selective contamination are required to explain the geochemical variations.

#### 2. Geology and sample suite

Detailed descriptions of the geology, petrography and geochemistry of the Miki Fjord macrodike and associated hybrids have been previously presented by Rosing et al. (1989) and Blichert-Toft et al. (1992); only a brief summary is given here. The macrodike is part of the East Greenland Palaeogene igneous province, which formed during opening of the North Atlantic Ocean basin. The province comprises mafic volcanics (e.g. Pedersen et al., 1997) cut by numerous Tertiary mafic to felsic plutons (e.g. Tegner et al., 1998), including the Skærgaard Intrusion. The Miki Fjord macrodike is one of a number of large dikes (typically ~500 m wide) in the vicinity of the Skærgaard intrusion (see also Naslund, 1989; Geist and White, 1994; Momme and Wilson, 2002) and outcrops over 15 km east of Skærgaard, intruding Precambrian gneiss overlain by Palaeogene basalt (Fig. 1). The Miki Fjord macrodike has a sub-ophitic texture and is dominated by ortho- and clinopyroxene, plagioclase and Fe-Ti oxides; olivine is rare (Blichert-Toft et al., 1992). The Precambrian basement hosting the macrodike is amphibolite-facies orthogneiss composed of plagioclase, alkali feldspar, quartz, hornblende, biotite, ilmenite, titanite, zircon and apatite and has yielded a metamorphic age of ~2.9 Ga (Leeman et al., 1976). The macrodike shows minimal compositional variation (e.g., 48-49.5 wt.% SiO<sub>2</sub>; 8-6.5 wt.% MgO) along much of its exposed length, except in areas close to the contacts with Precambrian rocks where felsic melts, formed by melting of adjacent basement lithologies, mingle with basaltic magma. A spectacular range of hybrid magmas are formed by interaction of these dissimilar magmas (Fig. 1). The zone of mingling along the contacts of the macrodike is 1 to 5 m wide and comprises visually homogeneous, fine-grained granophyre (74.9% SiO<sub>2</sub>) with an assemblage of quartz, alkali feldspar, plagioclase, apatite, zircon, titanite, magnetite and ilmenite and metre-size pillows of intermediate hybrid rocks with a diverse phase assemblage including pyroxene and plagioclase displaying striking resorption and zoning textures (a detailed description of the petrography of the hybrids is given by Blichert-Toft et al., 1992).

Sr and Nd isotopic investigations of the macrodike and surrounding hybrids show that the macrodike diabase has an isotopic composition  $(^{87}\text{Sr}/^{86}\text{Sr}_i\!=\!0.70392,\; \epsilon_{Nd(i)}\!=\!+6.5)$  (Table 1) broadly similar to the Skærgaard intrusion and other east Greenland Palaeogene basaltic magmas (Stewart and DePaolo, 1990; Blichert-Toft et al., 1992; Fram and Lesher, 1997). The compositions of these basaltic rocks contrast strongly with the Precambrian basement sample ( $^{87}$ Sr/ $^{86}$ Sr = 0.75596,  $\varepsilon_{Nd}$  = -37.5) and melt generated from it (granophyre;  $^{87}Sr_i=0.74882$ ,  $\varepsilon_{\rm Nd(i)} = -33.7$ ). A representative suite of these intermediate (hybrid) rocks, as well as the host granophyre, local gneiss and macrodike diabase, characterized by Blichert-Toft et al. (1992) for major elements, trace elements and Sr and Nd isotopes and collected from a single locality, serve as the sample suite for this study. Apart from a greyish-white hybrid (CL87-52H), interpreted by Blichert-Toft et al. (1992) as originally being granophyre based on field relations, granophyric texture and inclusions of more mafic hybrids, all the other hybrid rocks possess chemical and isotopic affinities that link them to the pristine diabase of the macrodike.

#### 3. Analytical methods

For each sample approximately 100 mg of rock powder was leached in hot 6 N HCl for 3 h to remove contamination from milling and weathering. The leachate was then discarded and the remaining sample washed several times with MQ water then attacked with concentrated HF and HNO<sub>3</sub> in a Parr Bomb for 48 h to ensure complete dissolution of resistant phases such as zircon. The samples were then dissolved using HNO3 and HCl, dried, and dissolved in a HBr-HCl solution. Pb was separated from the dissolved sample using anion exchange resin following methods described by Frei et al. (2002). The resultant Pb fraction was then diluted in 0.2% HNO<sub>3</sub>, doped with Thallium and analysed on an Axiom MC-ICPMS, using the measured  $^{205}\mathrm{Tl}/^{203}\mathrm{Tl}$  to correct for mass fractionation (assuming a natural value of 2.3889) using methods described by Baker et al. (2004). Replicate analyses of SRM981 carried out during the analysis yielded <sup>206</sup>Pb/  $^{204}\text{Pb} = 16.939 \pm 3$ ,  $^{207}\text{Pb}/^{204}\text{Pb} = 15.497 \pm 5$  and  $^{208}\text{Pb}/^{204}$  $^{204}\text{Pb} = 36.720 \pm 10 \ (n = 4)$  within error of a larger database of SRM981 analyses carried out in the laboratory using both Tl and double-spike techniques to correct for mass bias (Baker et al. 2004). These results are in good agreement with published values of Pb isotope standards determined elsewhere by double spike techniques (e.g. Thirlwall, 2002). Pb and U concentrations on rock powders were determined on separate dissolutions of unleached powders spiked with  $^{204}\text{Pb}$  and  $^{235}\text{U}$  tracers. Sample CL87-52F (host gneiss) was also separated into its major constituent minerals; a plagioclase feldsparrich separate (termed feldspar), an impure quartz-alkali feldspar separate (referred to as quartz + feldspar), biotite, amphibole, Fe-Ti oxides, zircon and apatite. Each mineral fraction (excluding zircon and apatite) was gently washed for two minutes in 6 M HCl. These clean mineral separates were dissolved and analysed unspiked. Apatite was dissolved directly in 6 M HCl and two separate zircon splits (large zircons (100–200  $\mu$ m) = zircon 1, small zircons (<100 μm) = zircon 2) were dissolved in high pressure Parr Bombs. Zircon was not abraded prior to dissolution. Zircon and apatite were spiked with a mixed 205Pb-235U tracer prior to dissolution. Pb and U concentrations and Pb isotope ratios for mineral separates were determined using the VG Sector 54 thermal ionisation mass spectrometer at Copenhagen University using methods described by Frei et al. (2002). Pb isotope ratios for zircon and apatite from CL87-52F are age corrected to 55 Ma (Table 1). Pb isotope ratios for whole rock and other mineral fractions are plotted in all diagrams with no age correction, given that the correction required is minor compared to the overall variation observed (less than the symbol size used, see Table 1).

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