Lu–Hf and Re–Os systematics of peridotite xenoliths from Spitsbergen, western Svalbard: Implications for mantle–crust coupling

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A R T I C L E   I N F O
Article history:
Received 5 January 2010
Received in revised form 4 June 2010
Accepted 7 June 2010
Available online 8 July 2010
Editor: R.W. Carlson

Keywords:
Lu–Hf isotope
Re–Os isotope
Spitsbergen
peridotite
delamination
Paleoproterozoic/Neoarchean

A B S T R A C T
The timing of sub-continental lithospheric mantle (SCLM) differentiation beneath Spitsbergen, western Svalbard, has been determined with spinel peridotite xenoliths using two complementary isotopic systems: Lu–Hf and Re–Os. The whole-rock Re–Os systematics (Re–Os errorchron, aluminochron, and Re-depletion age) define Paleoproterozoic/Neoarchean ages for isolation of the studied peridotites from the convecting mantle. We note that the age is independently supported by the Lu–Hf errorchron for the peridotite clinopyroxene grains, and average degree of melt depletion recorded in the samples. The obtained ages are indistinguishable from the oldest crustal ages reported in western Spitsbergen, implying that the mantle lithosphere in this area was stabilized at the same time as formation of the overlying crust. Our data suggest that the Spitsbergen lithosphere is unlikely to have undergone bulk lithospheric delamination since the tectonic transition from transpressional to transtensional which commenced in the Paleocene. We thus discount the delaminated and reactivated SCLM as a potential source for the Dupal-like enriched components in the Arctic upper mantle suggested by other studies.

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1. Introduction

Determining the age of lithospheric mantle offers critical information about development and stabilization of the continents and the genetic relationship between the mantle lithosphere and overlying crust. It also provides insights into mantle dynamics such as the potential contribution of delaminated and foundering mantle lithosphere to the source of geochemically enriched mantle components in derivative basaltic magmas. The unique geochemical characteristics of the Re–Os parent–daughter system provide insights into the timing of melt depletion events (hence lithospheric mantle stabilization) in peridotites, because partial melting fractionates Re from Os owing to Re being moderately incompatible while Os is strongly compatible. As melting lowers the Re/Os ratio of the residue, the time-integrated Os isotopic composition of that material experiences minimal to no radiogenic growth unlike the convecting mantle below. Furthermore, because the Os concentration in peridotites is 10 to 1000 times greater than in the derivative silicate melts or other metasomatic agents, the Os isotopic system in peridotites is thought to be more resistant to secondary overprinting compared with other radiogenic isotope systems (e.g., Sm–Nd and Rb–Sr) for which both parent and daughter elements behave incompatibly during mantle melting (e.g., Shirey and Walker, 1998).

Similarly, the Lu–Hf system has previously been shown to suffer no major effects from metasomatic processes and probably has a higher closure temperature than the Sm–Nd system, because (i) Hf has a considerably smaller diffusivity in silicate minerals than does Nd, and (ii) the relative difference in Hf concentration between melt and peridotites is lower than that in Nd concentration for the same materials (Pearson and Nowell, 2003; Choi et al., 2006, 2008; Bizimis et al., 2007; Wittig et al., 2007). Choi et al. (2008) demonstrated recently that the Lu–Hf system can be used to constrain the timing of lithospheric mantle differentiation and the study presented here supports that assertion.

Our study area, Svalbard Archipelago, is located on the northwesternmost edge of the Eurasian Plate, near the Knipovich Ridge and Gakkel Ridge in the North Atlantic and Arctic Oceans, respectively (Fig. 1A). Peridotite xenoliths brought to the surface by Quaternary volcanism on northwestern Spitsbergen, Svalbard, represent continental lithospheric mantle in the vicinity of oceanic spreading centers, and as such likely record the melt-extraction history and possibly even metasomatism in this tectonic setting. Previous work on these xenoliths mainly focused on the nature of metasomatism: processes, sources, and melt–fluid compositions (Amundsen, 1987; Ionov et al., 2007; Wittig et al., 2007).
1993, 1996, 2002a,b). The work showed that (1) residual Spitsbergen peridotite protoliths were affected by carbonate-rich silicate melt percolation, and (2) the metasomatic source is not directly related to the source regions of the host basalts. Recently, Goldstein et al. (2008) argued that delaminated and dispersed Svalbard lithospheric mantle during continental break-up might have been the source for the Dupal-like isotopic signature observed in basalts along the western Gakkel Ridge. Jung et al. (2009) reported deformation microstructures and water content of olivine in the peridotite xenoliths of the study area. We present here the first ever Lu–Hf and Re–Os isotopic determinations for the peridotite xenoliths from Spitsbergen. Our aim is to constrain the age of lithospheric mantle stabilization beneath the study area based on the two complementary systems, and to test the lithosphere delamination hypothesis.

2. Geological framework

Spitsbergen is the largest island in the Svalbard Archipelago of the Arctic Ocean, located midway between Norway and the North Pole (Fig. 1A). Tectonic reconstructions indicate that Svalbard was continuous with Greenland until the continental break-up and initiation of seafloor spreading between Norway and Greenland in the late Paleocene/early Eocene (Talwani and Eldholm, 1977; Johansson et al., 2005). The break-up of Laurasia along spreading
centers in the North Atlantic, Labrador Sea, and Eurasian Basin caused a transpressional tectonic regime on Svalbard during the Paleocene and Eocene, resulting in the creation of the Spitsbergen Transform Zone (Harland, 1969; Crane et al., 1982; Tuchschmid and Spillmann, 1992) (Fig. 1A). With cessation of sea floor spreading in the Labrador Sea in early Oligocene, the tectonic regime changed to transtension, leading to oblique extension in the Spitsbergen Transform Zone (Lena Trough), and increased spreading along the Knipovich Ridge (Taiwani and Eldholm, 1977; Hellebrand and Snow, 2003; Ljones et al., 2004). Sea floor spreading has propagated northward with time, resulting in linkage of the Arctic and North Atlantic at ca. 10 Ma (Eldholm et al., 1994; Blythe and Kleinspehn, 1998).

Two distinct phases of volcanism are known in Spitsbergen, the first consisting of Neogene lava flows, and the second, of Quaternary volcanic centers (Fig. 1B). Neogene lavas are largely subalkaline olivine tholeiitic basalts, whereas Quaternary volcanism has nepheline basanite/nepheline hawaiite rock composition. The volcanic centers are bounded by N–S trending faults which project towards the Yermak Plateau and parallel the Spitsbergen Transform Zone (Fig. 1A, B). These faults are considered to have been established or reactivated during continental fragmentation (e.g., Amundsen et al., 1987; Skjelkvåle et al., 1989). The volcanism is thus considered to be related to the opening of the eastern Arctic Basin and the Greenland Sea (Vogt et al., 1978; Feden et al., 1979), and the anomalously high heat flow on the Yermak Plateau, associated with reactivation of the Yermak hotspot (Crane et al., 1982; Tuchschmid and Spillmann, 1992).

Quaternary volcanic activity is found at three centers (Sverrefjell, Sigmufjell and Halvdanpengen) in the Bockfjorden area, NW Spitsbergen, along the trace of the Breibogen Fault zone (Fig. 1B). The fault Trends NNW–SSE and juxtaposes Mesoproterozoic marbles and gneiss—overprinted by Caledonian deformation—towards the west against Devonian sedimentary rocks in the east (Fig. 1B). The volcanic centers consist of basanite/hawaiite lava flows, pyroclastics and a hypabyssal intrusion. All centers contain abundant uppermost mantle and lower crustal xenoliths (Amundsen et al., 1987; Skjelkvåle et al., 1989). Samples for this study were collected from Sverrefjell, the richest xenolith locality.

3. Samples and petrography

Samples selected for this study include spinel lherzolites and spinel harzburgites. Table 1 gives a summary of the petrography, Mg# (=100Mg/(Mg+Fe)) of olivine, Cr# (=100Cr/(Cr+Al)) of spinel, and estimates of equilibration temperatures for the mineral assemblages in these samples. Mg#s of olivine vary from 90.3 to 92.3, and Cr#s of spinel from 9.7 to 60.0. The modal mineralogy of the xenoliths has been determined from calculations using bulk rock chemistry and mineral compositions. Sample SVF-26 shows direct evidence of modal metasomatism, where amphibole occurs as large equigranular grains, suggesting textural equilibrium with coexisting silicates. This sample has been included to help evaluate the effects of metasomatism on the Lu–Hf systematics. A few samples containing carbonate-bearing pockets were also collected to cover the more extreme end of the compositional spectrum. The pockets (up to 500 μm in size) consist of carbonate aggregates, second generation clinopyroxene and olivine, silicate glass, and relict grains of resorbed minerals of the host peridotite, which are usually located around spinel. Based on textural and geochemical data, previous studies (Ionov et al., 1996; Ionov, 1998) suggested that the Spitsbergen carbonate-bearing pockets were produced by reaction of carbonatite-rich mantle melt with the host peridotites shortly before their transport to the surface as xenoliths. Sulfides are present in most of the xenoliths, both as isolated inclusions within silicate phases and as interstitial components (Table 1, Fig. S1). Trapped inclusions in silicates are rounded blebs. Interstitial grains occur along grain boundaries, or as a tiny bleb trails cross cutting the silicate grains. The size of individual sulfide grains is variable (up to 200 μm in diameter); larger grains are generally enclosed in the silicate phases. Lherzolites have higher modal contents of sulfides than the harzburgites. The major element compositions of representative sulfides are given in Supplementary Table 1. They are monosulfide solid solution (MSS), exsolved into Ni-rich MSS to pentlandite with minor chalcopyrite.

Equilibration temperatures were estimated by two-pyroxene thermometer of Wells (1977), and Al–Cr exchange thermometer of Witt-Eickschen and Seck (1991) based on the Al–Cr partitioning between orthopyroxene and spinel coexisting with olivine. The

### Table 1

<table>
<thead>
<tr>
<th>Sample no</th>
<th>Rock type</th>
<th>Group Texture</th>
<th>Modes</th>
<th>Accessory minerals</th>
<th>Microstructural position of sulfides</th>
<th>Equilibration temperature</th>
<th>Al2O3 (wt.%)</th>
<th>Mg# in ol</th>
<th>Cr# in sp</th>
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<td>(sul) e (opx), i</td>
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</table>

Sample location: 12°21’50”E, 79°26’50”N.

Equilibration temperatures were estimated by two-pyroxene thermometer of Wells (1977), and Al–Cr exchange thermometer of Witt-Eickschen and Seck (1991). Modal compositions were calculated from whole-rock and mineral compositions by least-squares method.

Abbreviations: sp.lz = spinel lherzolite; sp.hz = spinel harzburgite; proto = protogranular; porphy = porphyroclastic; ol = olivine; opx = orthopyroxene; cpx = clinopyroxene; Sp = spinel; amp = amphibole; carb = carbonate-bearing pocket; sul = sulfide; e = enclosed in silicate grains; i = interstitial.

Mg# = 100Mg/(Mg + Fe).

Cr# = 100Cr/(Cr + Al).
equilibration temperatures range from 840 to 1020 °C (Table 1), which is within the range of temperatures (840 to 1170 °C) reported for Spitsbergen peridotites by Amundsen et al. (1987) and Ionov et al. (2002a). Amundsen et al. (1987) constructed a geotherm for northwestern Spitsbergen based on equilibrium P–T estimates for garnet websterite xenoliths, and temperature estimates for two-pyroxene granulate and spinel lherzolite xenoliths. Projection of the temperature estimates for the samples studied onto the Spitsbergen geotherm yields pressures of 7 to 11 kbar.

4. Analytical methods

The xenoliths range in size from 6 to 13 cm in their longest dimension. Samples for whole-rock analysis were prepared from the core of each xenolith, free of any visible surface weathering. The rock samples were first coarsely crushed to expose interior fragments of the xenolith using a tungsten-carbide pestle wrapped in paper to avoid contamination. They were then disaggregated using an agate mortar and pestle followed by grinding into a fine powder with an alumina ball mill. This produced 20 to 30 g of powder for each sample. Clinopyroxene is the major carrier of REE and possibly Hf in these samples. Samples were dissolved in a mixture of distilled 5% HF for 15 min, with an H2O rinse after each of these steps. They were then disaggregated using a magnetic separator to eliminate grains with dark inclusions, and hand picking under a binocular microscope to select only the optically clear grains for analysis. Prior to dissolution for the isotopic analyses, the minerals were washed in warm, distilled 2.5 N HCl for 15 min, and in warm distilled 5% HF for 15 min, with an H2O rinse after each of these steps.

Major-element concentrations have been determined by X-ray fluorescence (XRF) spectrometry on fused disks at Act Labs in Ontario, Canada (Supplementary Table 2). Rare-earth-element concentrations in clinopyroxene have been measured by inductively-coupled-plasma mass spectrometry (ICP-MS) at the Korea Polar Research Institute. Standards (JGB-1 and JR-1) have been analyzed with the unknown samples, and their values are reported to better than 1%. Matrix corrections were performed using internal standards. Total blanks averaged 0.04 ng for Nd, and 0.02 ng for Hf. The results are given in Table 2.

Re–Os isotopic analyses, including chemical separation and measurements, were performed at Japan Agency for Marine-Earth Science and Technology (JAMSTEC). Details of the analytical procedures appear in Suzuki et al. (2004) and Kato et al. (2005). Briefly, approximately ~0.5 g of pulverized whole-rock sample aliquots with 185Re and 186Os spikes were dissolved in Carius tube. This was followed by Os extraction with carbon tetrachloride and its purification by microdistillation. Re purification was carried out on anion exchange columns using AG 1 × 8 resin (100–200 mesh). Re and Os were loaded onto Pt filaments with Ba(NO3)2 as an activator. The Re–Os isotopic analyses were determined with a Thermoscientific TRITON®, in negative ion detection mode. Total blank levels were <2.0 pg for Re and <2 pg for Os with 187Os/188Os of ~0.17. Contribution of the blank to measured Os concentrations and 187Os/188Os were <0.1%. Precision of 187Re/186Re and 187Os/188Os measurements, based on analysis of an in-house standard over a period of several months, was better than 0.1% (1σ) and 0.3% (1σ), respectively. The results are given in Table 3.

Reproducibility based on replicate analyses of a high-concentration Os sample (SVF-41) was calculated to be 18% for Os concentrations, and 0.8% for the 187Os/188Os values. This is typical of platinum group element (PGE) data in general (e.g., Handler and Bennett, 1999; Alard et al., 2005). These variations can be attributed to the so-called nugget effect, which refers to heterogeneous distribution of PGE-bearing trace phases in a sample.

Table 2

Lu–Hf and Nd isotopic compositions for clinopyroxenes from Spitsbergen peridotite xenoliths, and for the host basanite.

<table>
<thead>
<tr>
<th>Sample no</th>
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<th>Hf (ppm)</th>
<th>176Lu/177Hf</th>
<th>176Hf/177Hf</th>
<th>2σ</th>
<th>εNd</th>
<th>143Nd/144Nd</th>
<th>2σ</th>
<th>εNd</th>
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<td>0.284655</td>
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<td>66.6</td>
<td>0.513075</td>
<td>22</td>
<td>8.5</td>
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<td>0.283779</td>
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<td>0.512916</td>
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<td>5.4</td>
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εNd and εHf are calculated with 176Hf/177Hf = 0.282772 and 143Nd/144Nd = 0.512638 for present-day chondritic Earth. sp.lz = spinel lherzolite; sp.hz = spinel harzburgite. n.d. = not determined.
Table 3
Re–Os isotopic compositions for Spitsbergen whole-rock peridotite xenoliths.

<table>
<thead>
<tr>
<th>Sample no</th>
<th>Rock type</th>
<th>Re (ppt)</th>
<th>Os (ppt)</th>
<th>187Re/188Os</th>
<th>187Os/188Os</th>
<th>2εγ</th>
<th>TEm (Ma)</th>
<th>TAMS (Ma)</th>
<th>γOs</th>
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All data are blank-corrected. The 187Os/188Os ratios are less than the 2-sigma reproducibility (0.6%) in the measurements of our Os standard.

5. Results

5.1. Whole-rock chemistry

The whole-rock Mg# values of the Spitsbergen peridotites studied range from 89.5 to 91.9 (Supplementary Table 2). Variations in representative major oxides (Al2O3 and CaO) as a function of MgO, good indicators of degree of depletion of the peridotites, are shown in Fig. 2A–B. Available data from previous study are shown for comparison. Spitsbergen peridotites are characterized by a wide range of compositional variation, extending from mildly depleted lherzolites (Al2O3 up to 3 wt.%), through harzburgites similar to typical abyssal peridotites, into the field of Archean cratonic harzburgites from East Greenland (Bernstein et al., 1998; Hanghøj et al., 2001) (Fig. 2A–B). The abundance and trends are compatible with the best-fit lines for world-wide spinel peridotite xenolith compositions, suggesting that the rocks are for the most part residues after variable degrees of basaltic melt extraction. We note that the samples containing carbonate-bearing pockets are not offset on the MgO vs. CaO plot, suggesting that the modal abundance of carbonates is not significant. The samples also do not display any significant FeO enrichment (Supplementary Table 2), even in the amphibole-bearing peridotite, ruling out amphibole formation from an evolved Fe-rich basaltic melt.

5.2. Rare earth element chemistry

Rare earth elements (REE) concentrations for the clinopyroxenes from the Spitsbergen peridotites are listed in Supplementary Table 3. Chondrite-normalized REE patterns are shown in Fig. 3. The heavy rare earth elements (HREE), such as Lu, which are moderately incompatible in clinopyroxene have abundances which correlate positively with indices of melt depletion (e.g., Al2O3) (Supplementary Tables 2 and 3; Fig. 4A), supporting an origin for the peridotites as residues after variable degrees of melt depletion. Meanwhile, the REE patterns for the clinopyroxene grains are variable from light rare earth element (LREE) depleted through spoon-shaped to LREE-enriched (Fig. 3A–C). These characteristics are not consistent with simple melt extraction, but instead reflect some overprinting by secondary metasomatism in the history of the xenoliths, as discussed in previous studies of the Spitsbergen peridotites (Ionov et al., 2002a,b). The samples (SVF-30, -49, -50, -53 and -61) retain residual characteristics inasmuch as they show LREE-depleted patterns and exhibit only very slight incompatible trace element enrichments. The degree of metasomatic overprinting thus increases from group a toward group b (SVF-5, −36, −47, −48, −51 and −52) to c (SVF-4, −9, −26, −41 and −42). Chondrite-normalized (Ce/Yb)N ratios vary from 0.3 to 1.3 for group a, 2.2 to 3.3 for group b (except SVF-51), and 3.9 to 20.5 for group c (Supplementary Table 3). Sample SVF-49 displays a fractionated HREE pattern, indicating that garnet might have been involved in the source.

5.3. Nd and Lu–Hf isotopes

The Nd and Lu–Hf isotopic compositions were obtained for clinopyroxene separates from 12 peridotites and the host basanite (Table 2). The Nd, Lu and Hf concentrations of clinopyroxene from Spitsbergen peridotites vary from 2.0 to 24.4 ppm, from 0.1 to 0.4 ppm, and from 0.2 to 1.1 ppm, respectively (Table 2; Supplementary Table 3; Fig. 4A–C), which lie within the range measured for other spinel peridotite xenoliths (e.g., Bizimis et al., 2003; Carlson et al., 2004; Choi et al., 2008). The clinopyroxene grains display a wide range in Nd isotopic compositions (143Nd/144Nd = 0.512693 to 0.513453; εNd = +1.1 to +15.9), and an extremely wide range in Hf isotopic compositions (176Hf/177Hf = 0.283368 to 0.286893; εHf = +21.8 to +149.3) and elemental ratios (176Lu/177Hf = 0.03 to 0.19). Host basanite has 143Nd/144Nd =...
characterized by strong Nd–Hf decoupling compared to the mantle array, whereas the host basanite plots within the field of oceanic basalts which define the array (Fig. 5A). It is remarkable that sample SVF-49 exhibits strongly depleted Hf isotopic composition (δHf = +149), in marked contrast to the present-day oceanic basalt mantle source. To test for reproducibility, we ran the sample in triplicate. The extreme Hf isotopic composition was reproduced within uncertainty (Table 2). Fig. 5A and 5B are the Sm–Nd and Lu–Hf isochron diagrams for the clinopyroxene separates from the xenoliths, respectively. Also plotted in the diagrams are available present-day DM (depleted mantle) values for reference, and Sm–Nd isotopic compositions for Spitsbergen peridotites (Ionov et al., 2002b). Group a samples have higher 147Sm/144Nd and 143Nd/144Nd ratios than the DM, in contrast to the group b and c samples which have lower values (Fig. 5A). Meanwhile, all but two samples have higher 176Lu/177Hf and 176Hf/177Hf ratios compared with DM (Fig. 5B). Sample SVF-49 has a highly elevated 176Lu/177Hf ratio, compared with the other samples (Fig. 5B). Therefore, the extremely depleted Hf isotopic composition reflects a time-integrated record of radiogenic Hf ingrowth associated with this high Lu/Hf. The elevated Lu/Hf ratio indicates that these clinopyroxene grains are likely to be the result of sub-solidus breakdown of garnet, which is consistent with the fractionated HREE pattern (Fig. 3A).

5.4. Re–Os isotopes

The Re–Os isotopic compositions were measured for ten whole-rock peridotites (Table 3). The Os concentrations of the Spitsbergen peridotites vary from 2.0 to 6.1 ppb, falling within the range for orogenic peridotites (global mean = 4.0±1.6 ppb) and cratonic peridotite xenoliths (4.2±2.7 ppb), but relatively higher than off-cratonic peridotite xenoliths hosted by alkali basalts (2.0±1.1 ppb) (Pearson et al., 2004) (Table 3). The Re concentrations vary from 0.03 to 0.22 ppb, which is less than the primitive mantle value of 0.28±0.08 ppb (McDonough and Sun, 1995) (Fig. 4d). This observation is consistent with variable Re loss through melt depletion. The xenoliths yield 187Os/188Os between 0.1087 and 0.1256, and 187Re/188Os ranging from 0.024 to 0.285. The Spitsbergen samples have lower 187Re/188Os and 187Os/188Os ratios (γOs = −16 to −3) than the estimates for the primitive upper mantle (PUM) (Meisel et al., 2001) (Fig. 6A, B), reflecting long-term isolation in a low Re/Os environment produced by ancient partial melting. Conventional Re–Os model ages (τRe–Os; Walker et al., 1989) — age of melt extraction occurred for individual peridotites from the intersection of two isotopic evolution paths defined by the present-day 187Os/188Os and 187Re/188Os ratios of the sample and PUM — range from 1.1 to 4.0 Ga with most falling within range of 1.6 to 2.0 Ga (Table 3). If the average chondritic mantle evolution curve (187Re/188Os = 0.40186, 187Os/188Os = 0.1270; Shirey and Walker, 1998) is used instead for the calculation, the estimated ages decrease by ca. 0.3 Ga. There appears to be no visible relation between the model ages and microstructural positions of the sulfides observed in thin section, classified as interstitial or enclosed (Tables 1 and 3; Supplementary Table 1; Fig. S1). Recent studies indicate that silicate-enclosed sulfides may represent residues of partial melting processes, but interstitial sulfides are often considered to be crystallization products of sulfur-bearing fluids (Alard et al., 2000; Pearson et al., 2002). These earlier studies report differences in major element compositions according to microstructural positions of the sulfides, such as relatively Cu–Ni enrichment in interstitial sulfides compared to silicate-enclosed ones. By contrast, no significant chemical difference is observed among these different types of sulfides in the Spitsbergen peridotites. Interstitial sulfides in Spitsbergen peridotites may not have formed by new growth but by recrystallization. In Fig. 6A, we have compared our data with published whole-rock abyssal peridotite Os isotopic compositions, noting that the 187Os/188Os ratio of sample SVF-41 falls well below any estimates for abyssal peridotites, reinforcing it as a representative of ancient residues of partial melting.
6. Discussion

6.1. Lu–Hf isotopic systematics

Previous studies (Ionov et al., 1996; 2002a,b) considered the possibility that carbonate-rich silicate melts percolating through residual peridotites might have produced the Spitsbergen peridotites which show ample evidence for modal and/or cryptic metasomatism. In order to test the hypothesis that Nd and Hf might be mobile during metasomatic overprinting, we have plotted the Nd and Hf abundances and isotopic compositions of clinopyroxenes from the Spitsbergen peridotites as a function of whole-rock Al₂O₃ concentration (Fig. 4B, C; Fig. S3A, B). The Nd abundances display a rough negative correlation with Al₂O₃ (Fig. 4B), and increase from group a through group b to c. Group a is composed of relatively fertile lherzolites, while group c is dominated by harzburgites. Therefore, this apparently contradictory pattern of stronger LREE enrichment in the most depleted samples can be attributed to metasomatic enrichment in LREEs of residual rocks formed by earlier partial melting events, which is very common feature observed in peridotite suites (e.g., Frey and Green, 1974). Group b and c samples have lower values of ¹⁴⁷Sm/¹⁴⁴Nd and ¹⁴³Nd/¹⁴⁴Nd than those of the DM in Fig. S2 further supports this notion. Meanwhile, the Hf concentrations show a general positive trend with Al₂O₃. The two group c samples (SVF-9 and -42), which have the lowest Al₂O₃ but the highest Nd concentrations, do not display coherent enrichment in Hf with Nd (Fig. 4B, C). Furthermore, the Hf concentration of a clinopyroxene from an amphibole-bearing peridotite (SVF-26) plots along the residual trend defined by the other samples (Fig. 4C). In addition, the clinopyroxene grains have a wide range in Nd isotopic compositions (εNd = +1.1 to +15.9) with a systematic decrease from group a through b to c. The Nd isotopic compositions display a good positive trend with Al₂O₃ (Fig. S3A). This strongly contrasts with the Hf isotopic compositions which show an extremely wide range in εHf (+21.8 to +149.3) without any systematic trend within the groups (Fig. 5A). In addition, most of the samples have elevated ¹⁷⁶Lu/¹⁷⁷Hf and ¹⁷⁶Hf/¹⁷⁷Hf ratios compared with the DM (Fig. 5B). These observations suggest that the Hf is much less susceptible to secondary overprinting by the carbonate-rich metasomatic melts than Nd. Therefore, we interpret the significant Nd–Hf isotopic decoupling observed in Fig. 5A to be the result of secondary metasomatism severely impacting the Sm–Nd system, but not the relatively more robust Lu–Hf system. Bizimis et al. (2003) showed that recent metasomatism of an ancient depleted peridotite through melt-rock reaction can generate Nd–Hf decoupling, similar to that seen in the Spitsbergen peridotites.

The ¹⁷⁶Hf/¹⁷⁷Hf ratios of lherzolites display a negative correlation with Al₂O₃ (Fig. S3B), supporting the notion that the Hf is immobile relative to Nd. Meanwhile, the Hf isotopic compositions of harzburgites (SVF-9, -42 and -52) plot outside the trend defined by lherzolites. It has been argued that orthopyroxene might be a significant repository for the HFSE (high-field-strength element) budget in anhydrous spinel peridotite (Rampone et al., 1991; Eggins et al., 1998; Bedini and Bodinier, 1999; Witt-Eickschen and O’Neill, 2001).
When we consider the partitioning value of Hf between clinopyroxene and orthopyroxene, $D_{\text{Hf}}^{\text{cpx/opx}} \approx 25$ in anhydrous spinel peridotite at ca. 900 °C (Eggins et al., 1998; Bedini and Bodinier, 1999; Witt-Eickschen and O’Neill, 2005), the contribution of orthopyroxene to the whole-rock budget for Hf in Spitsbergen lherzolites is estimated to be only up to $\approx 15\%$. Considering that the value of $D_{\text{Hf}}^{\text{cpx/amph}} \approx 1$ (Ionov et al., 2002a), then amphibole accounts for only 3% of the Hf in hydrous peridotite sample SVF-26. It appears that orthopyroxene and amphibole play only minor roles in the whole-rock Hf budget for the lherzolites. In harzburgites on the other hand, orthopyroxene can be a major repository of Hf. The high modal orthopyroxene/clinopyroxene ratio in Spitsbergen harzburgites results in the contribution of orthopyroxene ranging from 30 to 90% of the whole-rock Hf budget. Therefore, the offset observed in the correlation between Hf isotopic composition and whole-rock Al$_2$O$_3$ concentration (Fig. S3B) can be attributed to orthopyroxene control of the Hf budget in harzburgites.

The Spitsbergen peridotite clinopyroxenes define a positive correlation on a Lu–Hf isochron diagram (Fig. 5B). The samples yield an ‘errorchron’ age of 1.8 ± 1.1 Ga (2σ), an initial $\varepsilon_{\text{Hf}}$ of +28 ± 17 and a MSWD of 669 when the $\lambda_{176}\text{Lu}$ value of $1.865 \times 10^{-11}$ yr$^{-1}$ (Scherer et al., 2001) is used in the calculation. We interpret this Paleoproterozoic age to mark the time of stabilization of the mantle section by melt extraction, which will be supported by the Re–Os systematics in the next section below. Dating melting extraction by the Lu–Hf isochron method will be meaningful only if the original peridotites underwent melt extraction from an isotopically homogenous mantle source at the same time (or series of melting events closely spaced in time), and no Lu and Hf mobility has occurred since melt extraction. The large uncertainty might be due to a partial resetting by subsequent metasomatism, and/or multiple depletion events including source heterogeneity, although we do not have the resolution to quantify each factor precisely.

It is important to point out that the three harzburgite clinopyroxenes (SVF-9, -42 and -52) plot along the errorchron defined by lherzolite clinopyroxenes. Removing the three samples does not affect
the estimated errorchron age. The Lu/Hf ratio in orthopyroxene of spinel peridotite is expected to be higher than that of coexisting clinopyroxene (Eggins et al., 1998; Bedini and Bodinier, 1999; Witt-Eickschen and O'Neil, 2005), resulting in time-integrated growth of radiogenic Hf. If we assume that the Hf-isotopic equilibrium between clinopyroxene and orthopyroxene in the harzburgites was fast enough, the $^{176}\text{Hf}/^{177}\text{Hf}$ ratios of the coexisting clinopyroxenes might be significantly elevated compared with those of the lherzolite clinopyroxenes — considering the orthopyroxene control for the Hf budget in harzburgite; but this is clearly not the case. The Lu–Hf isotopic compositions of the three harzburgite orthopyroxenes are thus expected to be falling along the errorchron defined by clinopyroxenes with higher values. That is, the Lu–Hf system seems to be unaffected by diffusional equilibration between pyroxenes. What we can say from our results is that the closure temperature for Lu and Hf diffusion in clinopyroxene of spinel peridotite might be quite high, on the order of ~900 °C or perhaps even higher.

6.2. Re–Os isotopic systematics

The Spitsbergen peridotites define generally positive correlations on the Re–Os isochron diagram (Fig. 6A), and Re vs. $\text{Al}_2\text{O}_3$ diagram (Fig. 4D). Sample SVF-36 with an extreme composition on the Re–Os isochron, contains carbonate-bearing pockets (Table 1), and has elevated Re concentration for its $\text{Al}_2\text{O}_3$ (Fig. 4D). The scattered data thus can be attributed to open-system behavior of Re during recent mantle metasomatism (e.g., Walker et al., 1989; Reisberg and Lorand, 1995; Carlson et al., 1999; Gao et al., 2002). However, while it is not possible to rule out some effect of metasomatism on the $^{187}\text{Os}/^{188}\text{Os}$ isotopic compositions, all samples possess sub-chondritic values (Fig. 6A) suggesting that such effects have been minimal. Even sample SVF-36 has sub-chondritic Os isotopic composition, suggesting that the elevated Re content was acquired only recently. The Os concentrations do not define a meaningful correlation with $\text{Al}_2\text{O}_3$ (Fig. S3C). The Os in spinel peridotites is considered to reside primarily in discrete sulfides (e.g., Shirey and Walker, 1998; Handler and Bennett, 1999; Burton et al., 2000). The Re also resides in a sulfide component in the spinel peridotite, with some control by clinopyroxene (Hart and Ravizza, 1996; Handler and Bennett, 1999; Burton et al., 2000). The scatter observed in Os concentrations (Fig. S3C) may simply reflect nugget effects caused by heterogeneous distribution of trace phases, possibly exsolved from monosulfide solid solutions during cooling (Handler and Bennett, 1999; Alard et al., 2002; Pearson et al., 2004). Meanwhile, $^{187}\text{Re}/^{188}\text{Os}$ ratios define a relatively good correlation with $\text{Al}_2\text{O}_3$ concentrations (Fig. S3D), except for sample SVF-36, further suggesting that Re and Os isotopic compositions have not been significantly disturbed by subsequent metasomatism. Our observation is reminiscent of a previous study by Handler and Bennett (1999) on peridotite xenoliths from S.E. Australia, in which PGEs and Re were not significantly mobilized during interaction with carbonate melts.

If the Re–Os isochron is forced through PUM, apart from sample SVF-36 the regression, the Spitsbergen peridotites yield an errorchron age of 2.1 ± 1.1 Ga, an initial $\gamma_{\text{Re}}$ of 0.0 ± 2.3 and an MSWD of 0.278 when the $^{187}\text{Re}$ value of 1.666 × 10$^{-11}$ yr$^{-1}$ (Shirey and Walker, 1998) is used in the calculation (Fig. 6A). We note that this apparent age is comparable to the independent estimates obtained from the Lu–Hf errorchron (Fig. 5B), even though there is a debate for the true value for PUM (Walker et al., 2002).

Further confirmation comes from determination of the Re-depletion age ($T_{\text{RD}}$) defined by Walker et al. (1989) This approach provides a model age to compare the Os isotopic composition of the xenolith at the time of host lava eruption to a primitive mantle growth model by assuming that the xenolith has no Re. If all of the Re was removed at the time of melting, then the $T_{\text{RD}}$ age should equal the $T_{\text{MA}}$ age. $T_{\text{RD}}$ ages are always minimum ages. They provide good approximations for the time of melting in highly refractory peridotites, but severely underestimate the melting age of more fertile samples. The two most refractory samples analyzed for Re–Os isotopes, SVF-36 and SVF-41, which both contain less than 1% of $\text{Al}_2\text{O}_3$, yield $T_{\text{RD}}$ ages of 2.24 and 2.71 Ga, respectively (Table 3).

Alternatively, Re mobility can be overcome by plotting the Os isotopic compositions against an immobile element that exhibits a similar degree of incompatibility to Re during melting, such as $\text{Al}_2\text{O}_3$, CaO and HREE (Reisberg and Lorand, 1995; Handler et al., 1997). If the data display a positive correlation, then the $^{187}\text{Os}/^{188}\text{Os}$ of the y-intercept (Reisberg and Lorand, 1995) or the $^{187}\text{Os}/^{188}\text{Os}$ present at the lowest likely $\text{Al}_2\text{O}_3$ concentration (e.g., 0.7 wt.% $\text{Al}_2\text{O}_3$) (Handler et al., 1999) can be considered as the initial ratio. The time of melt depletion can be determined by comparing the initial ratio to a primitive mantle growth model. Spitsbergen samples display a relatively good correlation ($R^2 = 0.78$) in the $\text{Al}_2\text{O}_3$ vs. $^{187}\text{Os}/^{188}\text{Os}$ pseudo-isochron (aluminachron) in Fig. 6B. The trend passes through the hypothetical PUM, suggesting that they reflect melt extraction processes from an essentially primitive mantle source. We also observe good trends between Os isotopic compositions and Mg# ($R^2 = 0.76$, CaO ($R^2 = 0.68$), and indicators of melt depletion such as Cr# in spinel ($R^2 = 0.60$) (Tables 1 and 2; Supplementary Table 2). Assuming that the correlation between $^{187}\text{Os}/^{188}\text{Os}$ and $\text{Al}_2\text{O}_3$ records a single melting event or series of melting events closely spaced in time, then the extrapolation of the pseudo-isochron to $\text{Al}_2\text{O}_3 \approx 0.6$ wt.% gives a model age of 2.2 ± 0.5 Ga (Fig. 6B), which is within error of the apparent ages defined by Lu–Hf and Re–Os isochrons above. Sample SVF-41, which is the most refractory and has the lowest $\text{Al}_2\text{O}_3$ (0.6 wt.%) — in fact falling below the lower limit (ca. 0.7 wt.%) at which it has been suggested Re isotope values approach zero during partial melting of the upper mantle (Handler et al., 1997) — has the highest Os concentration in the entire suite. This sample is characterized by a significantly lower $^{187}\text{Os}/^{188}\text{Os}$ value of 0.1096, defining a Neoarchean $T_{\text{MA}}$ of 2.7 Ga, which is nearly equal to the $T_{\text{MA}}$ age of 2.9 Ga (Table 3; Fig. 6B). The ages are supported by duplicate analyses (Table 3) of this sample. Collectively, the Paleoproterozoic/Neoarchean ages are confirmed by two independent methods. We thus interpret the computed age range as the time when this piece of mantle was accreted to the continental lithosphere beneath Spitsbergen.

6.3. Mantle–crust coupling and implications for the host basanite mantle source

It is generally acknowledged that the mean compositions of subcontinental lithospheric mantle sampled as xenoliths, garnet concentrates, and peridotite masses have evolved from highly depleted Archean cratonic mantle to more fertile Phanerozoic mantle, probably associated with declining mantle temperature or a change in the mechanism of continental lithosphere generation or both (e.g., Boyd, 1989; Griffin et al., 1999, 2003; Pearson et al., 2004; Carlson et al., 2005). This evolution has been illustrated in a plot of olivine content against Mg# of olivine (Fig. 7), and a whole-rock Mg# against Cr# (Fig. S4). The fields of archon, proton and tecton, as defined by Griffin et al. (1999) (i.e., lithospheric mantle that experienced its last tectonothermal event at >2.5 Ga, 2.5–1.0 Ga, and <1.0 Ga, respectively, inferred from the age of the overlying crust) are shown for comparison in Fig. 54. Many xenoliths from Archean cratons are characterized by their higher Mg#s compared with off-cratonic samples, which may be related to a higher average degree of partial melting, possibly at greater depth. Spitsbergen peridotites are generally more depleted than the peridotites produced in Phanerozoic time, and plot in the field for hypothetical Proterozoic cratons. This result independently supports the Paleoproterozoic/Neoarchean ages determined with Lu–Hf and Re–Os systematics above.

Svalbard has been interpreted to have originated by amalgamation of several different terranes during large-scale transient faulting in the waning stages of the Caledonian Orogeny (e.g., Harland, 1997;
Lyberis and Manby, 1999; Johansson et al., 2005). Based on comparison of pre-Caledonian and Caledonian tectonic histories, Svalbard can be subdivided into the western and eastern terranes along the Billefjorden Fault Zone (Fig. 1B) (e.g., Gee et al., 1994; Johansson et al., 2005). The western province of Svalbard is considered to be derived from northern Greenland or Ellesmere Island in the Canadian Arctic, whereas eastern Svalbard is thought to have originated from eastern Greenland (Fig. 1A) (Johansson et al., 2005, and references therein). The Paleoproterozoic/Neoproterozoic geological event has hitherto not been recognized in the western province of Svalbard. However, Bernard-Griffiths et al. (1993) showed that protoliths of eclogite tectonic lenses in a Caledonian high-pressure metamorphic complex from central western Spitsbergen yield a mid-Paleoproterozoic age (igneous zircon; 2121±50 Ma U–Pb zircon discordia upper intercept age; Sm–Nd model age for the corresponding whole-rock sample = 2.17 Ga with εNd(T) = +5.2). In northern Greenland and southeastern Ellesmere Island, Paleoproterozoic and Neoarchean gneiss complexes have been investigated by numerous conventional and SHRIMP zircon U–Pb age determinations together with whole-rock Sm–Nd isotopic analyses (Frisch, 1988; Frisch and Hunt, 1988; Nutman et al., 2008). Rough agreement between the ages for melt depletion of the Spitsbergen peridotites and the oldest crustal rock, suggests that stabilization of the lithospheric mantle beneath Spitsbergen may have been linked with formation of the overlying crust.

On the Sm–Nd isochron diagram (Fig. S2), the Spitsbergen peridotites define an errorchron age of 620 ± 260 Ma (2σ), an initial εNd of +9.8 ± 6.1 and a MSWD of 18. This may reflect partial resetting of the Sm–Nd system by the Caledonian orogeny, but we do not preclude simple mixing of residual radiogenic protolith with enriched metasomating agent(s). The metasomatic components are unlikely to be related with the host basalts, considering that (1) the host basalts have intermediate Sm–Nd isotopic compositions compared to the Spitsbergen peridotites (Fig. S2), and (2) the Lu–Hf isotopic composition of the host basanite falls off the trend defined by the peridotites on the Lu–Hf isochron plot (Fig. 5B). That is, the host basalts do not possess end-member isotopic characteristics. This finding is consistent with previous work for Sr–Nd isotopic systematics of the Spitsbergen peridotites and the host basalts (Ionov et al., 2002b).

Recently, Goldstein et al. (2008) showed that the western Gakkel Ridge volcanism was derived from an enriched mantle with Dupal-like characteristics, and that the Spitsbergen host basalts form an enriched end-member for the Gakkel basalts on Sr–Nd–Pb isotopic diagrams. The authors argued that the Svalbard lithospheric mantle might have been delaminated, and dispersed into the convecting Arctic asthenosphere during the separation of Svalbard and Greenland, resulting in formation of an enriched component with a Dupal-like signature. This idea supposes that the host basaltic volcanism originated by partial melting of the Spitsbergen lithospheric mantle. We present here some evidence which refutes this idea. First, we note that the host basanite has a Hf isotopic composition that is coupled to its Nd isotopic composition, therefore plotting along the mantle array, within the field for oceanic island basalts on the Nd–Hf isotopic plot (Fig. 5A). This is distinctly different from the peridotite xenoliths which exhibit significant Nd–Hf decoupling, even when we consider only the weighted mean (143Nd/144Nd = 0.51309±0.00015, and 176Hf/177Hf = 0.28420±0.00062) of the measured values.

Furthermore, the host basanite contains comparatively less radiogenic Hf isotopic compositions than any of the peridotite xenoliths (Fig. 5A). This observation implies that a genetic relationship between the xenoliths and the host basaltic volcanism is weak at best. In fact, the host basalts display less radiogenic 206Pb/204Pb ratios than the Spitsbergen xenoliths at a given 207Pb/204Pb ratio on the 206Pb/204Pb vs. 207Pb/204Pb diagram (Fig. 4 in Ionov et al., 2002b), which further supports our notion. Most importantly, the Re–Os and Lu–Hf age data we have presented here suggest that the ancient Paleoproterozoic/Neoproterozoic lithospheric mantle persists beneath western Spitsbergen despite the tectono-magmatic reactivation and change in tectonic regimes from transpressive to transtensive since the Paleocene (e.g., Tuchschild and Spillmann, 1992; Ljones et al., 2004). It should be stressed that there is no meaningful correlation between temperature and whole-rock compositions or isotopic compositions for the Spitsbergen samples (Tables 1–3; Supplementary Tables 2 and 3), which implies that the lithospheric mantle is not stratified with respect to composition, and that the ancient fragments are not relics preserved locally at relatively shallow depth. These lines of evidence suggest that it is unlikely for detached and recycled bulk portions of the lithospheric mantle beneath the Svalbard Archipelago to have become the enriched source for the western Gakkel Ridge volcanism.

If we simply define ‘lithospheric thickness’ as the depth where the geotherm intersects the mantle adiabat (∼1280 °C) — although its definition is somewhat arbitrary — lithospheric thickness derived from the xenoliths-defined Spitsbergen geotherm (Amundsen et al., 1987) is estimated to be about 25 kbar, corresponding to a thickness of ∼85 km. The samples we have studied are estimated to have originated at depths equivalent to ∼7 to 11 kbar of pressure. This may leave a potential for deeper portion of the lithosphere to be delaminated or mechanically eroded during extension of the Lena Trough and become the enriched magma source in the shallow convecting mantle. Although our understanding of the nature of lithospheric root is not yet crystal clear, we may expect, if any, the deepest lithospheric mantle beneath Spitsbergen to have relatively little contrast in geochemistry compared to the asthenosphere. If this is the case, input of deeper part of the lithospheric mantle into the Arctique magma sources is hard to identify. Furthermore, it is unreasonable to expect lateral variation of the magma sources from distance from the continental margin. Overall, the evidence does not support reactivated subcontinental lithospheric mantle (SCLM) as the prevalent enriched end-member for the Dupal-like signature in the Arctic Ocean Basin.

Recent Re–Os studies have shown that some abyssal peridotites record melting events older than the age of the ridge they are dredged from, on spatial scales of individual sulfide grains within a single sample or even in bulk-rock analyses (Brandon et al., 2000; Harvey et al., 2006; Liu et al., 2008). These inherited, ancient partial melting effects have also been reported in peridotites brought to the surface by lavas in the Hawaiian plume, in forearc peridotites, and in ophiolites (Parkinson et al., 1998; Bizimis et al., 2007; Pearson et al.,

![Fig. 7. Modal weight percent vs. Mg# (= 100 Mg/(Mg + Fe)) of olivine from Spitsbergen peridotites. Data sources: fields of Archean lherzolites and harzburgites, and Phanerozoic lherzolites from Griffin et al. (1999); oceanic peridotite trend from Boyd (1989).](image-url)
2007), which were taken as evidence for a heterogeneous upper mantle. The refractory nature of the melt-depleted peridotites—that is its higher solids temperature than fertile mantle—might result in long-term survival in the convecting mantle (Harvey et al., 2006; Bizimis et al., 2007; Liu et al., 2008), although much work is needed in the future in order to fully understand the isolation mechanism. This observation raises an issue that the correlation observed in the Spitsbergen peridotites (Fig. 6B) may represent simple mixing between relatively fertile and ancient, refractory mantle preserved in the convecting mantle for billions of years. For this to be the case, some assumptions are necessary, such as that (1) the peridotites have been only recently emplaced at lithospheric depths (ca. 840–1020 °C/7–11 kbar), and (2) a significant amount of lithospheric erosion occurred before the emplacement at current levels. However, there is no available seismological data to support this model yet. Furthermore, we note that the Paleoproterozoic/Neoarchean age has been only recently emplaced at lithospheric depths (ca. 840 °C) and that the Paleoproterozoic/Neoarchean age has been independently supported by the Lu–Hf systematics and geochemistry of the peridotites. We thus interpret it as the lithospheric mantle formation age beneath Spitsbergen. Combined Lu–Hf and Re–Os isotope systematics are therefore required to adequately estimate the stabilization age of the lithospheric mantle.

7. Conclusions

(1) The Paleoproterozoic/Neoarchean age of SCLM incorporation in Spitsbergen has been confirmed by several independent methods: Lu–Hf and Re–Os isotopic systematics, and geochemistry of the peridotite xenoliths.

(2) The long-term survival of the SCLM, despite protracted tectonomagmatic reactivation in this area, argues against delaminated mantle lithosphere as a significant source of geochemically enriched mantle components in the Arctic region.

(3) The Lu–Hf system can be used to date mantle melting events recorded in peridotite xenoliths, in combination with the Re–Os system.

(4) Carbonate-rich metasomatic melts have severely impacted the Sm–Nd system of the Spitsbergen residual peridotites, but not the Lu–Hf and Re–Os system.

(5) Clinopyroxene grains from Spitsbergen peridotite are characterized by strong Nd–Hf decoupling on isotopic correlation diagrams, whereas the host basanite plots within the field for oceanic basalts. The host basanite volcanism is not likely to have been derived by partial melting of the Spitsbergen lithospheric mantle.

Acknowledgements

Funding for this project has come from Polar Academic Program (PAP), Korea Polar Research Institute to SHC. This work was supported in part by Grant-in-Aid from JSPS to KS. Insightful reviews by Laurie Reisberg and John Lassiter greatly improved the manuscript. We would like to thank B.K. Park, K.H. Kim, Y.J. Jwa, M. Kusakabe, K. Nagao, and S.B. Park for their assistance with field work in 2007, and A.V. Andronikov, C. Stefano, H. Yamamoto, M.K. Choo, and J.U. Seo for assistance with analytical work.

Appendix A. Supplementary data

Supplementary data associated with this article can be found in the online version, at doi:10.1016/j.epsl.2010.06.013.

References


