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Feldspar–Ti-oxide metasomatism in off-cratonic continental and oceanic upper mantle

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Abstract

Metasomatism is responsible for enrichment of lithospheric mantle in incompatible elements. The most common metasomatic products in mantle xenoliths and peridotite massifs worldwide are amphibole and mica providing mineral hosts for alkalis, Ba, Nb. In contrast, xenoliths of spinel peridotite entrained in basaltic rocks in southern Siberia (Russia) and the Kerguelen islands (Indian ocean) have metasomatic aggregates of alkali feldspar and Ti-rich oxides (rutile, armalcolite, ilmenite) that commonly replace Al-spinel and orthopyroxene or make up cross-cutting veins. Importantly, the feldspar-rich aggregates also replace amphibole and mica formed by earlier metasomatic episodes. Armalcolite has not been previously reported in off-cratonic peridotite xenoliths. The unusual mineralogical composition of these metasomatic products defines a specific type of mantle metasomatism, distinct from those commonly attributed to H₂O-rich fluids, carbonate melts or Fe–Ti-rich silicate melts. This type of metasomatism operates both in continental and oceanic mantle and may be related to alkali-rich fluids/melts with low water activity (probably due to high CO₂/H₂O ratios). Electron microprobe analyses have shown that the metasomatic rutile and armalcolite may have up to 4% of Nb₂O₅ and ZrO₂. Feldspar analysed by laser ablation ICP-MS typically has high contents of light REE, Rb, Ba, Sr. Overall, the feldspar-rich metasomatic aggregates may be strongly enriched in incompatible trace elements, with HFSE largely hosted by the Ti-rich oxides. Precipitation of this mineral assemblage can lead to unusual fractionations among incompatible elements both in the metasomatic assemblage and associated fluids. © 1999 Published by Elsevier Science B.V. All rights reserved.

Keywords: lithosphere; mantle; metasomatism; armalcolite; rutile; feldspar group; trace elements

1. Petrography

Metasomatism is responsible for enrichment of lithospheric mantle in incompatible elements. The most common metasomatic products in mantle xenoliths and peridotite massifs worldwide are amphibole and mica [1,2] providing mineral hosts for alkalis,

Ba, Nb [3]. However, feldspar and Ti-rich oxides have been found in about half of some 90 spinel lherzolite xenoliths examined in thin sections from seven alkali basaltic occurrences in the northern Sikhote-Alin (Far East of Russia) [4] and Hamar-Daban Range south of Lake Baikal [5], and in three harzburgite xenoliths from the Kerguelen islands [6]. Their mode of occurrence is similar at all localities. The feldspar (fs) and oxide minerals in association with secondary olivine (ol) make up fine-grained aggre-

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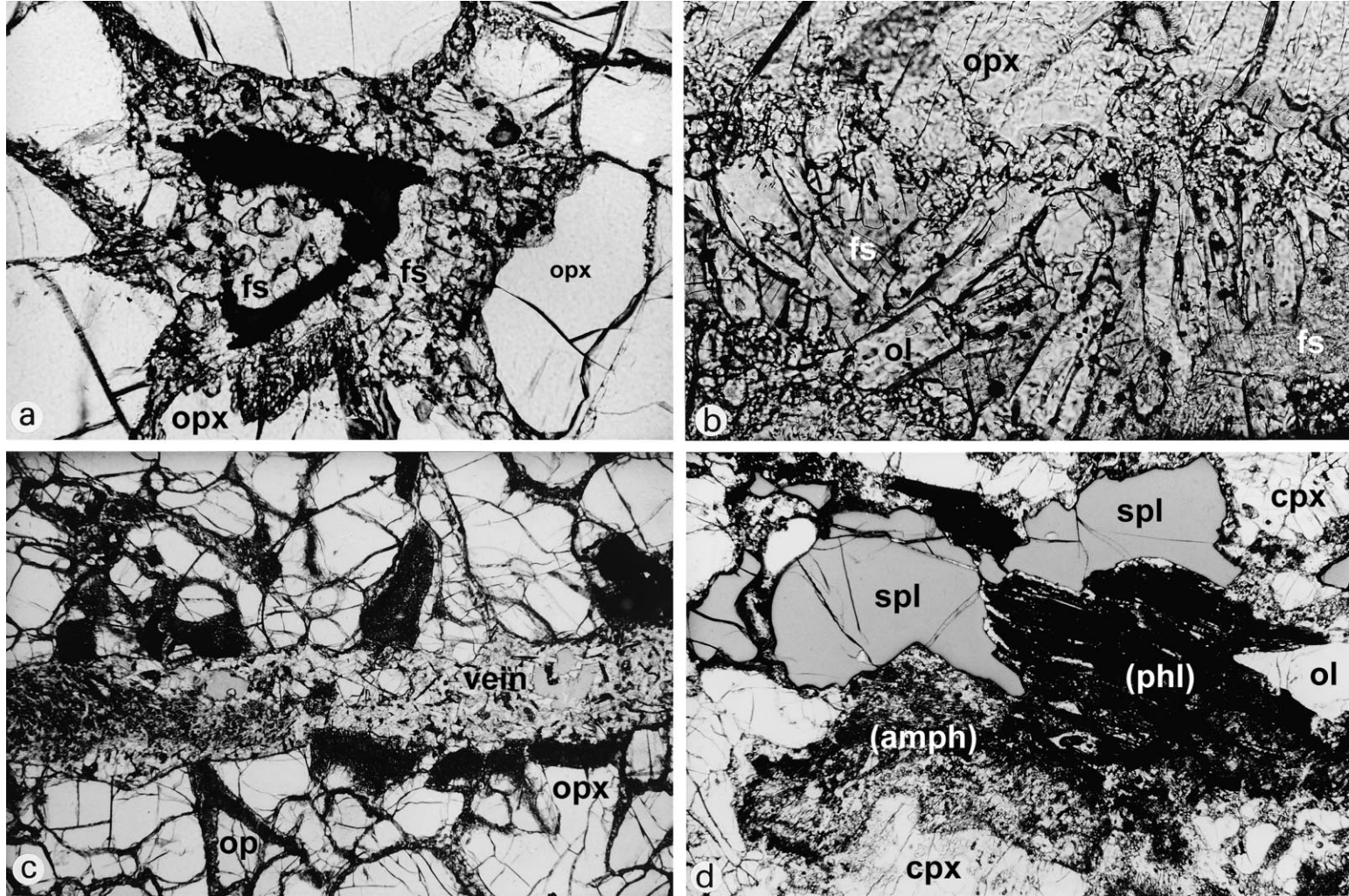


Fig. 1. Photomicrographs of xenoliths in transmitted light: (a) fine-grained olivine–feldspar (*fs* = feldspar, *ol* = olivine) aggregate replacing a corroded spinel (*spl*) grain and orthopyroxene (*opx*) (Sikhote-Alin, field width 1.2 mm); (b) contact area of a vein (bottom) of olivine, interstitial feldspar (dark) and opaque oxide needles replacing orthopyroxene (Kerguelen, field width 0.25 mm); (c) a cross-cutting feldspar-rich vein with needle-shaped Ti-oxides in a spinel lherzolite, note widespread replacement of orthopyroxene and spinel near the vein (Sikhote-Alin, field width 5 mm); (d) replacement of amphibole (*amph*) and mica (*phl*) in a coarse amphibole–phl–cpx–spl vein in a spinel lherzolite by fine-grained feldspar-rich aggregate (Sikhote-Alin, field width 5 mm).

gates in reaction zones and interstitial veins adjacent to corroded spinel (spl) and orthopyroxene (opx) (Fig. 1a,b). Microstructures and mineral chemistry suggest the reaction: $\text{Al-spl} + \text{opx} + \text{fluid} = \text{fs} + \text{ol} + \text{Cr-spl} \pm \text{Ti-oxides}$. Some xenoliths also contain thin (≤ 2 mm) cross-cutting veins made up of prismatic feldspar crystals and subordinate subhedral olivine, clinopyroxene (cpx) and oxide minerals (Fig. 1c). No silicate glass has been observed in the metasomatic aggregates and veins. The feldspar-bearing patches or veins have sharp contacts with host volcanic rocks. Whole rock feldspar contents are low ($\ll 1\%$) in the Kerguelen xenoliths and may reach 2–3% in the Siberian xenoliths.

Amphibole and mica occur in a few xenoliths from Sikhote-Alin as relics in fine-grained feldspar-rich patches (Fig. 1d). One Kerguelen xenolith has large mica grains rimmed by feldspar. No unaltered amphibole or mica was found in xenoliths from the Siberian sites. Textural evidence from the Siberian xenoliths suggests that amphibole and mica are much more likely to be replaced by the feldspar-rich metasomatic products than coexisting spinel and orthopyroxene. Fig. 1d shows an almost complete replacement of large (2–3 mm) amphibole and phlogopite grains (in a coarse amph–phl–cpx–spl vein in a spinel lherzolite) whereas adjacent spinel grains have only thin alteration rims. Other xenoliths have similar feldspar-rich patches that contain no relics of pre-existing minerals and may have formed by complete replacement of amphibole or mica. In some cases the fine-grained patches have plate-like cross-sections that may have been inherited from replaced mica grains. It is not likely that these patches were formed after other minerals because spinel and orthopyroxene in these xenoliths show only minor metasomatic reactions at grain rims. The textural evidence suggests that amphibole and mica are likely to break down at the onset of the metasomatism, by contrast with ‘anhydrous’ minerals that show more limited metasomatic alterations probably proportional to the amount of fluid added.

2. Mineral composition

Minerals were analysed for major and minor elements by electron microprobe (EMP). Equilibration

temperatures estimated by the Ca-opx method [7] are 900–1010°C for the Siberian xenoliths and 1010–1100°C for the Kerguelen xenoliths. The feldspar is generally alkali-rich but exhibits broad compositional variations. The $(\text{K} + \text{Na})/(\text{K} + \text{Na} + \text{Ca})_{\text{at}}$ ratio ranges from 0.3 to 1.0; the overall range for K_2O is 0.2–11.2%. Significant variations in the K/Na/Ca ratios may exist from sample to sample, within individual xenoliths and even between nearby feldspar grains. The K-rich feldspar typically has high TiO_2 contents (0.2–0.6%).

Four Ti-rich oxide minerals are identified in the xenoliths by EMP analysis: rutile, armalcolite, Ca–Cr armalcolite and ilmenite (Table 1, Fig. 2a). Armalcolite ($\text{Fe,MgTi}_2\text{O}_5$) was first discovered in lunar high-Ti basalts and subsequently recognized in metasomatized xenoliths from kimberlites and related rocks erupted through old cratonic mantle where it commonly occurs together with rutile, ilmenite, mica and K-amphibole, but not with feldspar [9,8,10]. We emphasize that the Ti-oxides in the samples we studied are associated with alkali feldspar, but not with amphibole and mica, and the metasomatic mineral assemblage in these xenoliths is distinct from the assemblages in which the Ti-oxides occur in the cratonic xenoliths.

The Ti-rich oxide minerals seem to be extremely rare in peridotite xenoliths outside cratonic terrains. In particular, armalcolite has not previously been reported in basalt-borne xenoliths. Rutile was only found (together with ilmenite) as microphases on spinel surfaces in association with mica, though textural relationships between the Ti-oxides and mica have not been documented [11]. Accessory ilmenite was reported in rare metasomatized xenoliths [12]. It is possible, however, that fine-grained Ti-oxides have been overlooked in some xenolith suites.

The Ti-rich oxides in xenoliths from Siberia and Kerguelen are compositionally very similar (Table 1, Fig. 2), both to each other and to those in xenoliths from kimberlites [8], e.g. they may contain significant Cr, Zr, Nb. Rutile, armalcolite and Ca–Cr armalcolite may have up to 4% of Nb_2O_5 and ZrO_2 . Ilmenite contains $<0.4\%$ Nb_2O_5 and ZrO_2 . EMP analyses with detection limits of 0.02–0.05% found no Th, Sr, Ba or Ce in the Ti-oxides. Host volcanic rocks contain only ilmenite that has low $\text{Mg}/(\text{Mg} + \text{Fe})$ ratios and is Cr-free.

Table 1
Electron microprobe analyses of Ti-rich oxide minerals (wt.%)

Oxide	Ilmenite	Ca–Cr armalcolite		Armalcolite		Rutile
	9513-37	GM501	XD-1	GM501	9513-8	9513-7
TiO ₂	54.14	65.89	67.60	70.65	61.74	91.67
Al ₂ O ₃	0.04	1.35	0.88	1.39	0.01	0.11
Cr ₂ O ₃	1.78	9.30	10.21	2.93	1.62	0.92
∑FeO	30.88	4.12	7.58	11.90	23.07	0.62
MgO	10.71	7.05	3.82	10.07	5.20	0.00
CaO	0.03	2.73	4.24	0.34	0.03	0.05
ZrO ₂	0.22	2.99	1.59	0.38	2.47	3.10
Nb ₂ O ₅	0.11	0.39	0.25	0.24	3.51	1.24

Sample location: 9513-7, -8, and -37, Sikhote-Alin; XD-1, Hamar-Daban; GM501, Kerguelen.

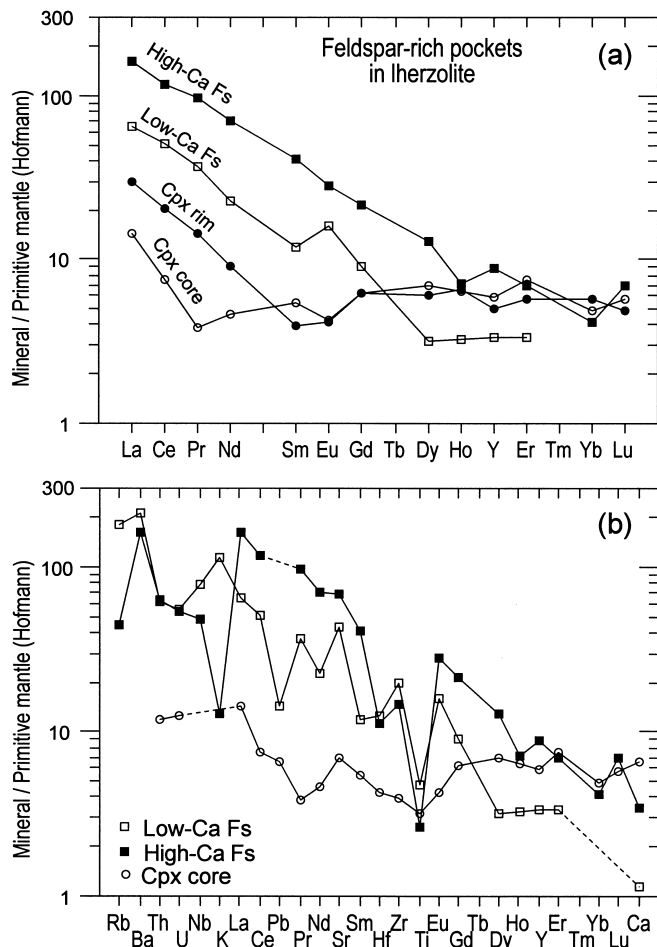


Fig. 3. Primitive mantle-normalized [14] abundance patterns for interstitial feldspar (*Fs*) grains with high Ca (filled boxes) and low Ca (open boxes) and for clinopyroxene (open circles, core; filled circles, rim) from lherzolite xenolith 9513-37 (Sikhote-Alin): (a) REE and Y, (b) extended trace element diagram.

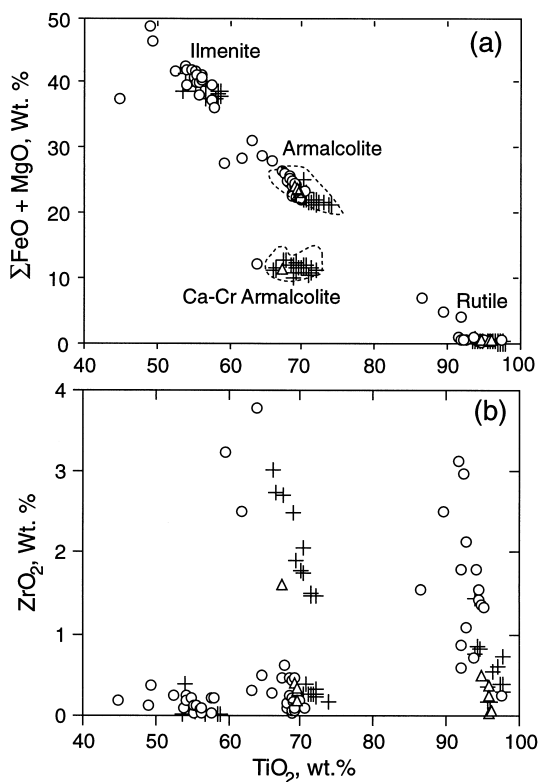


Fig. 2. Compositional variations of Ti-rich oxide minerals in xenoliths from Sikhote-Alin (circles), Hamar-Daban (triangles) and Kerguelen (crosses): (a) TiO_2 vs. $\Sigma\text{FeO} + \text{MgO}$; (b) TiO_2 vs. ZrO_2 . Dashed lines show approximate contours of fields for kimberlitic and lunar armalcolites and Ca–Cr armalcolites [8].

Trace elements in feldspar and primary clinopyroxene were determined in polished rock sections by laser ablation microprobe inductively coupled mass spectrometry (LAM ICP-MS) with detection limits of ≤ 0.1 ppm and precision of $\pm 5\%$ [13]. Clinopyroxene in the majority of the Siberian xenoliths we analysed (e.g., 10 out of 15 samples from Sikhote-Alin) is depleted in light rare earth elements (REE) relative to heavy REE. Feldspar is enriched in light over heavy REE and typically has much higher contents of LREE and similar (for Ca-rich feldspar) or somewhat lower contents of HREE and Y than coexisting clinopyroxene (Fig. 3a and Fig. 4a). By comparison, estimates of cpx/feldspar partition coefficients suggest that plagioclase (pl) should have lower REE and Y contents than coexisting clinopyroxene, with $\text{cpx/pl} \geq 100$ for HREE and Y [15].

The two minerals do not appear to be in equilibrium, with feldspar trace element patterns reflecting an enrichment episode whereas the primary clinopyroxene in many xenoliths has retained a signature of an earlier depletion event. In some xenoliths clinopyroxene is LREE-enriched, with higher LREE in the rims than in the cores (Fig. 3a). The LREE-enrichment in the rims might have been produced by diffusional exchange with metasomatic media during feldspar formation, but could also be due to an earlier metasomatic event.

Feldspar in cross-cutting veins has positive Eu anomalies (Fig. 4) whereas interstitial feldspar has no conspicuous Eu anomaly (Fig. 3). Because Eu commonly has higher fs/melt partition coefficient ($D_{\text{Eu}}^{\text{fs/melt}}$) than adjacent REE [15] this is an important distinction reflecting differences in the origin of the two types of feldspar. The interstitial feldspar is inferred to form by a metasomatic reaction triggered by infiltration and in situ crystallization of alkali-rich fluids without REE fractionation. The Eu-enriched vein feldspar may have crystallized under open system conditions from melts flowing in conduits consistent with lower $\text{Mg}/(\text{Mg} + \text{Fe})_{\text{at}}$ ratios of vein olivine (0.72–0.84) than for olivine associated with interstitial feldspar (0.87–0.92). The $D_{\text{Eu}}^{\text{fs/melt}}$ is known to vary significantly depending on oxygen fugacity. f_{O_2} estimates for xenoliths from Sikhote-Alin, including feldspar-bearing peridotites (0 to +0.4 log units relative to the FMQ buffer), fall in the middle of the range typical of asthenospheric mantle and MORB glasses (0 ± 1.5 log units relative to FMQ [16]) assumed by Rampone et al. [15] for $D_{\text{Eu}}^{\text{fs/melt}}$ estimates. Distinct f_{O_2} values in fluids producing interstitial and vein feldspar in spatially related samples do not appear to be likely.

The feldspar is generally rich in Rb, Ba and Sr, but the contents of highly incompatible elements vary broadly, both between nearby feldspar grains and between different samples. Ca-rich feldspar has higher contents of REE, Y and Sr and lower Rb than Ca-poor feldspar in the same xenolith (Fig. 3). Feldspar grains with abundant oxide inclusions yield high contents of Nb, Zr, Hf (Fig. 4b) apparently hosted by Ti-rich oxides; these grains also have high contents of REE, Th, U, indicating that the feldspar may contain microinclusions of yet unidentified phases rich in these elements.

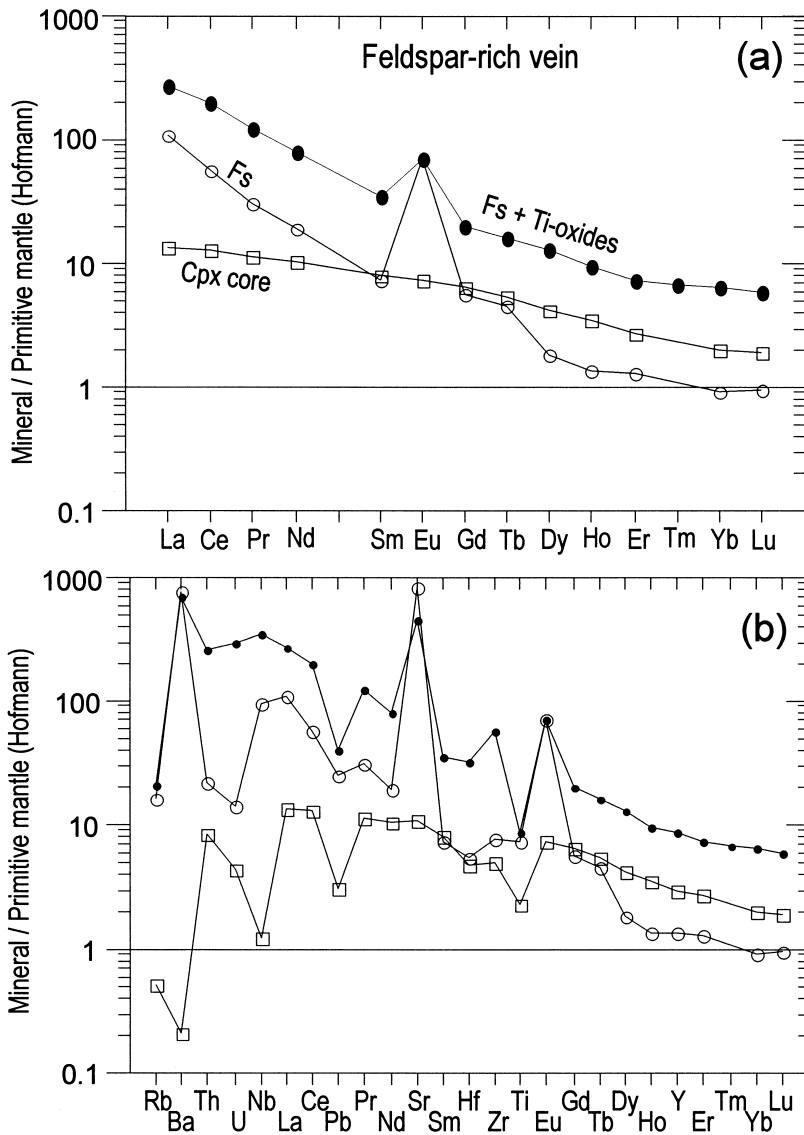


Fig. 4. Primitive mantle-normalized [14] REE (a) and trace element (b) abundance patterns for feldspar in a vein (circles) and for clinopyroxene from the host harzburgite (boxes) in xenolith GM-92-501 from Kerguelen. Individual oxide minerals were not analysed because they are smaller than the pits produced by laser beam (30–50 μm), but feldspar grains with Ti-oxide inclusions (filled circles) yield much higher contents of REE, Zr, Hf, Nb, Th and U than inclusion-free grains (open circles).

The variations in trace element contents in feldspar, Ti-rich oxide minerals and clinopyroxene and in modal abundances of the metasomatic minerals in the xenoliths may produce a variety of whole-rock trace element patterns. The feldspar-oxide pockets, veins and whole-rock xenoliths from

Sikhote-Alin and Kerguelen commonly show consistent enrichment in incompatible elements from heavy to light REE, with moderate to high Zr, Hf, Nb, Th, U, Ba, Rb, K. In contrast, some feldspar-bearing Hamar-Daban xenoliths are enriched in K and Rb, but not in other incompatible elements [5].

The feldspar in the Siberian xenoliths has Rb/Cs ratios of 140–180 which are significantly higher than estimates for the bulk silicate Earth (28), island-arc lavas (20–30), upper crust (20), lower crust (60) and averages for kimberlites (30), mid-ocean-ridge basalts (MORB) and ocean island basalts (OIB) (80) [17]. It is not clear whether the Rb/Cs ratios in the feldspar are similar to those in its parental metasomatic agent. This assumption may be valid for the interstitial feldspar which probably formed by complete in situ crystallization of fluid/melt, and considering that the xenoliths contain no other alkali-bearing minerals. In any case, the Rb/Cs ratios in the feldspar from the Siberian xenoliths are nearest to those in MORB and OIB, and may indicate that the alkali-rich metasomatic fluids were derived from an asthenospheric mantle source rather than from subduction-related or crustal sources.

3. Discussion

The formation of the alkali feldspar and associated Ti-rich oxides is likely to be a relatively recent event because they are not in textural and chemical equilibrium with host peridotites. That process, however, is distinct from partial melting of hydrous minerals, spinel and pyroxenes in xenoliths shortly before and during their entrainment in host magma [18,19] because the feldspar-bearing xenoliths do not contain silicate glass. These rocks are also different from plagioclase peridotites in massifs and basalt-borne xenoliths that contain K-poor plagioclase (but no Ti-oxides) which commonly coexists with amphibole and is related to injection of tholeiitic melts at low-pressure conditions [5,15,20].

Petrographic evidence from the Siberian xenoliths suggests that the introduction of even small amounts of the metasomatic fluid which precipitates feldspar and Ti-oxides results in a rapid breakdown of amphibole and mica, probably because they are not stable in the presence of such a fluid. The evidence for breakdown of presumably hydroxyl-bearing amphibole and mica and their replacement by ‘anhydrous’ feldspar and Ti-oxides indicates low water activity in the fluid responsible for metasomatism. Experimental results in the peridotite–CO₂–H₂O system at 10–15 kbar suggest that subsolidus dehydration of

amphibole and mica may take place if CO₂/(CO₂ + H₂O) ratios exceed 0.6–0.8 [21,22]. Reaction of carbonated alkali magma with spinel peridotite [23] may also produce feldspar and oxides and residual CO₂-rich melt or fluid at *P–T* conditions when carbonates are not stable in shallow mantle. Water activity may be very low in concentrated supercritical NaCl–KCl solutions [24].

In summary, our data suggest that specific fluid compositions may bring about modal metasomatism in spinel–lherzolite facies mantle, both continental and oceanic, in which trace element enrichment is largely hosted by alkali feldspar and Ti-rich oxides rather than amphibole and mica. Precipitation of this mineral assemblage can lead to unusual fractionations among incompatible elements both in the metasomatic assemblage and associated fluids. In particular, Ti-oxides may cause decoupling of high field strength elements (HFSE) from other elements in migrating fluids resulting in HFSE anomalies in mantle rocks and melt source regions. This type of metasomatism however is not likely to be responsible for negative Nb–Ta anomalies in arc magmas because it probably occurs only in shallow mantle (limited by the stability of alkali feldspars in peridotite [5]) and in fluid environments with low water activity. It is not clear why the feldspar–Ti-oxide metasomatism is widespread in a few Siberian xenolith suites from different tectonic settings (rift zone, active continental margin). Similar metasomatic products may be overlooked in other xenolith suites worldwide because they are fine-grained and tend to form thin interstitial films and small pockets.

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