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# Spinel inclusions in olivine of peridotite xenoliths from TUBAF seamount (Bismarck Archipelago/Papua New Guinea): evidence for the thermal and tectonic evolution of the oceanic lithosphere

Received: 25 May 2000 / Accepted: 12 July 2000

**Abstract** Olivine in spinel peridotite xenoliths from the Bismarck Archipelago northeast of Papua New Guinea, which were transported to the surface by Quaternary basalts, shows spinel inclusions up to 25 µm long and 200 nm wide. These inclusions mainly occur as inhomogeneously distributed needles and subordinately as octahedral grains in olivine of veined metasomatic peridotites as well as peridotites without obvious metasomatism. The needles very often occur in swarms with irregular spacing in between them. Similar spinel inclusions in olivine have only previously been reported from ultramafites of meteoritic origin. Composition and orientation of the spinel inclusions were determined by transmission electron microscopy (TEM) and analytical electron microscopy (AEM). Both the needles and the grains display a uniform crystallographic orientation in the host olivine with  $[001]_{\rm Ol}//[1\overline{10}]_{\rm Spl}$  and  $(100)_{\rm Ol}//(111)_{\rm Spl}$ . The needles are elongated parallel [010] in olivine, which is the same in all olivine grains. As these needles have no relation to the metasomatic sections in the peridotite, it is concluded that they are primary features of the rock. Although the composition of the spinel needles is often very similar to the large chromian spinel octahedra in the matrix, the small octahedral spinel inclusions in olivine are in part Mg-rich aluminous spinel and sometimes almost pure magnetite. The spinel needles are suggested to have formed by exsolution processes during cooling of Al- and Cr-rich, high-temperature olivine during the initial formation of the lithospheric mantle at the mid-ocean ridge. The Al-rich spinel octahedra probably formed by the breakdown of an Al-rich phase such as phlogopite or by metasomatism, whereas the magnetite was generated by oxidizing fluids. These oxidizing fluids may either have been set free by dehydration of the underlying, subducted plate or by the Quaternary magmatism responsible for the transport of the xenoliths to the seafloor.

#### Introduction

Inclusions are a common feature in olivine of mantle peridotites and in magmatic olivine; numerous types of symplectitic exsolutions of spinel, ortho- and clinopyroxene are described in literature (Bell et al. 1975; Arai 1978; Putnis 1979; Moseley 1984; Puga et al. 1999). Similar observations of needles in olivine are reported from several rock types. Needle-shaped lamellae of chromian spinel and clinopyroxene were detected in olivine from the Iwanai-Dake peridotite mass in Japan (Arai 1978), and lamellae of kirschsteinite (CaFeSiO<sub>4</sub>) were found in olivine of the angrite meteorite (Mikouchi et al. 1995). Magnetite lamellae in olivine and clinohumite were described from ultrahigh-pressure metamorphic rocks from the Dabie Shan in China (Zhang et al. 1999) and needles of magnetite and clinopyroxene were reported from intrusive rocks from Greenland (Markl et al. 2000). Coupled exsolutions of chromian spinel and CO<sub>2</sub> (and sometimes native carbon) may occur in olivine from mantle xenoliths (Green and Gueguen 1983; Green 1985). Most of these inclusions are interpreted as exsolutions because of decompression, cooling or oxidation processes. In natural rocks, inclusions commonly form polyphase assemblages consisting of spinel, pyroxenes and/or a fluid phase. Descriptions of single-phase chromian or aluminous spinel inclusions in olivine are rare and limited to chondritic meteorites (Ashworth 1979). We report here on tiny spinel needles and octahedral grains embedded in olivine of mantle peridotite xenoliths from the TUBAF seamount in the Bismarck Archipelago, northeast of Papua New Guinea. Electron

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Editorial responsibility: J. Hoefs

microprobe and transmission electron microscope (TEM, AEM) investigations reveal the composition and the crystallographic orientation of these inclusions in the host olivine and allow an interpretation of their formation.

## **Geological setting**

The Bismarck Archipelago northeast of the island of Papua New Guinea comprises the islands of the New Britain arc-trench system, New Ireland, New Hanover, Bougainville and the Solomons (Fig. 1). The archipelago is underlain by the Bismarck microplate, which was generated in a mid-ocean ridge (MOR) environment (McInnes et al. 1999). In this area, the Pacific plate in the NE underwent subduction below the Bismarck microplate in the SW along the so called Manus-Kilinailau trench. The subduction, which was accompanied by strong calc-alkaline volcanism in Oligocene and Miocene, was stopped ~15 Ma ago, when a collision of the Ontong-Java plateau with the trench occurred (Coleman and Kroenke 1981). The stalling of the subduction zone resulted in the development of a subduction reversal leading to the formation of the New Britain trench, on which a northward subduction of the Solomon Sea microplate under the South Bismarck microplate is presently taking place (see Fig. 1; McInnes and Cameron 1994).

The island of Lihir belongs to the NW-SE trending Tabar-Lihir-Tanga-Feni volcanic island chain extending for a distance of more than 250 km northeast of New Ireland. Volcanic activity started ~3.6 Ma ago in the New Ireland fore-arc region and continues until now (Licence et al. 1987; McInnes 1992; Rytuba et al. 1993). The high-K calc-alkaline volcanism can be related to lithospheric extension along NE-trending faults (Taylor 1979; Steward and Sandy 1988) and incorporates alkali-olivine basalts, trachybasalts, trachyandesites, ankaramites and foid-bearing mafites (Wallace et al. 1983; Kennedy et al. 1990). These melts were generated as a consequence of the thinning of the New Ireland basin lithosphere and subsequent decompression. The magma genesis is strongly influenced by the stalled subduction along the Manus-Kilinailau trench. Metasomatizing agents from the underlying subducted Pacific plate penetrated the overlying mantle segment and induced the formation of veined mantle (McInnes and Cameron 1994; McInnes et al. 1999, 2000; Franz et al. 2000).

In the course of two cruises of the research vessel RV Sonne, 12 submarine volcanoes were mapped south of Lihir Island (Herzig

et al. 1994, 1998). One of these volcanoes, the so called TUBAF seamount, is made up of pristine pyroclastic deposits and characterized by the absence of pelagic sediments indicating a relatively recent eruption. The ankaramitic magmatites of this volcano contain numerous fresh mantle xenoliths, which were sampled by video-guided grab. In many cases, the mantle peridotites gave evidence for vivid metasomatism in the form of pyroxene-, hornblende-, and phlogopite-bearing veins. A detailed description of the mineral assemblages, the vein-forming reactions and the geochemistry of these rocks has recently been performed (Franz et al. 2000).

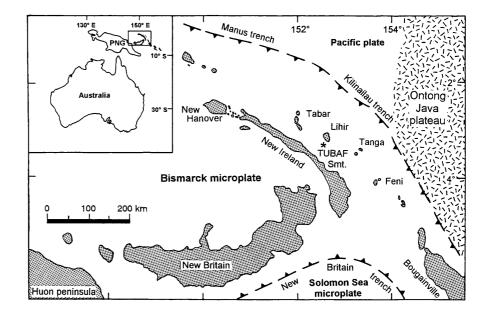
## Sample preparation and analytical details

Microprobe analyses were performed using a JEOL Superprobe with five spectrometers at the University of Freiberg. Major and minor elements were determined at 15 kV acceleration voltage and a beam current of 20 nA with counting times of 20 s for Si, Al, Mg, Ca, Sr, Ba and K, and 30 s for Fe, Ni, Na, Cr, Mn and Ti. Ca, Al and Cr in olivine were determined with an acceleration voltage of 20 kV, a beam current of 55 nA, and a counting time of 300 s on the peak. The standard sets of the Smithsonian Institute (cf. Jarosewich et al. 1980) and MAC were used for reference.

The TEM specimens were prepared from standard thin sections without a cover glass. The areas of interest were removed from the thin section, glued onto a copper grid and finally thinned with an ion beam (5 kV, 12° tilt of the ion guns). After thinning, the specimen were slightly coated with carbon to prevent charging.

TEM and AEM were carried out in a Philips CM200 operated at 200 kV. The electron source was a LaB<sub>6</sub> filament. The TEM is equipped with an EDAX X-ray analyzer with ultrathin window. The spectra were corrected for absorption and fluorescence and analysis totals normalized to 100%. The spectra were acquired in the scanning mode using a nominal beam diameter of 4 nm. To minimize irradiation damage the beam was scanned over an area of typically  $40 \times 30$  nm in size. The counting time was 120–180 s, which gives sufficiently high counts to keep the error from counting statistics low. The k<sub>AB</sub> factors were determined from measuring at least 20 different areas of the standard. Specimen thickness, necessary for absorption correction was determined by electron energy-loss spectroscopy (EELS) using the zero-loss peak and the whole spectrum (Egerton 1996). The EEL spectra were recorded with a Gatan GIF. The total error (error from kAB-factor determination + the error from the counting statistics) is  $\sim$ 3% relative (rel.)

**Fig. 1** Map of the Bismarck Archipelago with the position of the TUBAF seamount; *inset* shows the location of the area northeast of Papua New Guinea (PNG)



for MgO and SiO<sub>2</sub> and 7% rel. for FeO in olivine. For low elemental concentrations the total error might exceed 50% rel.

Element mapping of the spinel inclusions was performed with the GIF, applying the jump ratio or elemental mapping method of the Gatan Digital Micrograph software package.

#### **Results**

Petrography, mineral chemistry and thermobarometry of the xenoliths

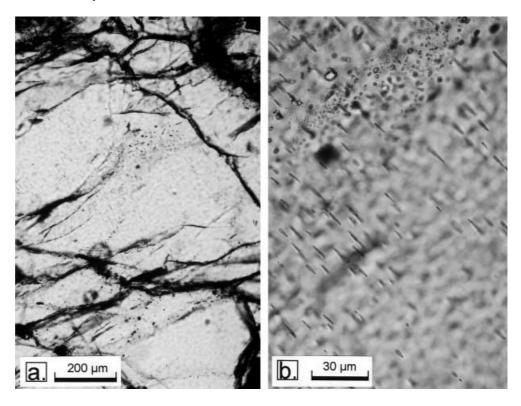
Most of the mantle xenoliths are pristine spinel and plagioclase peridotites with coarse-equant textures following the classification of Harte (1975). Especially the spinel peridotites reveal numerous, needle-shaped spinel crystals and inhomogeneously distributed spinel octahedra in olivine (Fig. 2). The most common are needles with a typical length of 5–25 µm and 50–200 nm width, which are concentrated in some areas of a grain, thus forming swarm-like pattern with the same crystallographic orientation. Other areas of the same grain may contain only a few needles or they are completely absent. The spinel needles often occur in the center of the grains and have no relation to microfractures in the olivine grains or trails of fluid inclusions. They are found in veined metasomatized samples as well as in ultramafic xenoliths without obvious metasomatic alteration. Two typical ultramafic xenoliths - one veined metasomatized lherzolite and one harzburgite without obvious secondary alterations – were selected and investigated with the microprobe and TEM.

Sample 56-2P is a granular spinel harzburgite with minor amounts of clinopyroxene. It mainly consists of

olivine with Fo<sub>90-91</sub>, which displays a bell-shaped zonation pattern with significantly decreasing Ca-content from core (120 ppm) to rim (1 ppm). Orthopyroxene with  $X_{Mg} = Mg/(Mg + Fe^{2+})$  -values of 0.9, contains low concentrations of Al<sub>2</sub>O<sub>3</sub> (1.4 wt%) and Cr<sub>2</sub>O<sub>3</sub> (0.3 wt%) and has a wollastonite component (Wo) of 0.9–1.2 mol%. Numerous exsolution lamellae of clinopyroxene are found in the orthopyroxene. Mapping of the orthopyroxene using a 50-µm beam of the microprobe leads to significantly higher concentrations of Al, Cr and Ca than spot analyses with a beam diameter of 1 μm (Table 1). Clinopyroxene is Cr-diopside with Wo<sub>43-44</sub>, En<sub>53-54</sub> and Fs<sub>3</sub> (calculation after Lindsley 1983), which contains plenty of exsolution lamellae of orthopyroxene. Charge balance constraints point to minor amounts or to an absence of ferric iron in clinopyroxene. Large, intergranular spinel octahedra with diameters of 0.1–0.9 mm are Cr-rich with  $X_{Cr} = Cr$ (Cr + Al)] values of 0.49–0.53 and  $X_{Mg}$  values of 0.59– 0.61 (Table 1). Towards the rim of the spinel, a decrease in chromium and ferric iron at the expense of aluminum is observed.

As evident from the zonation pattern in olivine as well as the exsolution lamellae in pyroxene, a distinct cooling occurred in the xenolith. Pyroxene thermometry using the T BKN calibration of Brey and Köhler (1990) yields temperatures of ~820 °C, using point analyses of the pyroxenes while integrated analyses (mapping of the crystals using a 50-μm beam diameter of the microprobe) reveal temperatures of ~880 °C. Similarly, the spinel—olivine Fe–Mg-exchange geothermometer of Ballhaus et al. (1991) yields temperatures of ~770 °C for the core and 700 °C for the rim section of the large

Fig. 2 a Microphotograph of an olivine grain with spinel needles Needles are accumulated in the central part of the grain and nearly absent at the rim (upper left corner and right side of the photograph). b Enlargement of a showing spinel needles near a trail of fluid inclusions. Note that there is no relation between the needles and the trail



**Table 1** Representative analyses of spinel, olivine and pyroxene from sample 56-2P. Fe<sup>3+</sup> estimated according to the method of Droop (1987). AEM analytical electron microscopy;

Spl (wt%)	56-2P Al-Spl	56-2P Lamella	56-2P Lamella	56-2P Lamella	56-2P Lamella	56-2P Lamella	56-2P Spl	56-2P Spl	Ol (wt%)	56-2P Ol I avg.	Cpx (wt%)	56-2P Cpx I	Opx (wt%)	56-2P Opx I
	Core AEM	1 AEM	2 AEM	3 AEM	4 AEM	5 AEM	Core	Rim EMP		(n = 108)  EMP		Core EMP		Core EMP
SiO <sub>2</sub>	5.30	2.50	1.80	3.60	2.10	1.60	0.00	0.00	SiO <sub>2</sub>	40.40	SiO <sub>2</sub>	54.10	SiO <sub>2</sub>	56.88
$Al_2O_3$	59.80	26.30	24.90	23.40	23.70	26.70	25.02	27.00	$A_2O_3$	0.00	Al <sub>2</sub> O <sub>3</sub>	1.63	$Al_2O_3$	1.80
$Cr_2O_3$	0.20	40.30	40.40	38.70	41.10	37.60	41.43	39.86	$Cr_2O_3$	0.00	$Cr_2O_3$	0.73	$Cr_2O_3$	0.45
$M_{\rm gO}$	25.20	0.00 14.50	3.30 16.10	5.72 17.90	5.18 15.90	3.29 17.20	12.32	12.71	MgO CaO	0.01 0.01	$M_{\rm gO}^{\rm rc_2O_3}$	0.00 17.46	F <sub>2</sub> O <sub>3</sub> MgO	33.35
CaO	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	MnO	0.00	CaO	23.60	CaO	0.71
MnO	0.00 9.50	0.00 16.30	0.00 13.73	0.00 13.16	0.00 14.24	0.00 12.14	0.13 16.33	0.12 16.01	Nio Nio	8.65 0.38	MnO FeO	0.06 1.72	MnO FeO	5.67
ZnO	0.00	0.00	0.00	0.00	0.00	0.00	0.14	0.35	Total	98.66	$Na_2O$	0.09	$Na_2O$	0.00
NiO Total	100.00	0.00	0.00 100.23	0.00 100.47	0.00 100.22	0.00 $100.53$	0.10 98.45	0.08 98.90			$\mathbf{K}_2\mathbf{O}$ Total	0.00 99.44	$\mathbf{K}_2\mathbf{O}$ Total	0.01 99.06
Cations $(O = 4)$									Cations (O	0 = 4	Cations (	(9 = 0)	Cations (O	(9=
i Si		0.074	0.053	0.106	0.063	0.047	0.000		i S:	0.988	i Zi	1.968	i Z:	1.974
- T	0.000	0.000	0.000	0.000	0.000	0.000	0.002		I V	0.000	- F	0.001	= <del>Z</del>	0.001
, C	0.004	0.947	0.948	0.898	0.970	0.869	1.015		j;	0.000	Ċ.	0.021	, Cr	0.012
re. Mo	0.000	0.000	0.0 /4	0.082	0.0/1	0.117	0.068		g S S	1.839	Te.	0.000	Μο	0.000
Ca	0.000	0.000	0.000	0.000	0.000	0.000	0.000		Mn	0.000	Ca	0.920	Ça Ca	0.027
$\mathop{\mathrm{Mn}}_{\scriptscriptstyle{ m Po}^{2}}^{+}$	0.000	0.000	0.000	0.000	0.000	0.000	0.003		공 :Z	0.177	$\mathop{ m Mn}_{{ m Fe}^{2^{+}}}$	0.002	$\mathop{ m Mn}_{{ m Fe}^2^+}$	0.005
Zn	0.000	0.000	0.000	0.000	0.000	0.000	0.003	0.008	Total	÷	S S	0.006	Na Sa	0.000
Total	2.997	2.991	3.000	3.000	3.000	3.000	3.000	3.000	CaO (ppm) Cr <sub>2</sub> O <sub>3</sub> (ppr Al <sub>2</sub> O <sub>3</sub> (ppn	n) 151 n) 15 n) 20	N Total	3.988	<b>N</b> Total	3.983
$X_{\mathrm{Mg}}$	0.83	0.61	89.0	0.71	0.67	0.72	0.57	0.58	End-member	LS	End-mem Lindsley (	bers After	End-members	rs
$X_{Cr}$	0.00	0.51	0.52	0.53	0.54	0.49	0.53	0.50	$Fe_2SiO_4$	8.74		(2271)	$\mathrm{Fe}_2\mathrm{Si}_2\mathrm{O}_6$	8.57
P (kbar) T BBG	15.0 758	15.0 713	15.0 877	15.0 936	15.0 862 0.33	15.0 971	15.0 725	15.0 719	$Ca_2SiO_4$ $Ni_2SiO_4$	0.02 0.37	Wo: En:	43.30 53.70	Mg <sub>2</sub> Si <sub>2</sub> O <sub>6</sub> Mn <sub>2</sub> Si <sub>2</sub> O <sub>6</sub>	89.79 0.26
Delta $\log f_{\mathrm{O2}}$	1	_	-0.18	0.00	-0.23	0.54	-0.22	-0.32	Mn <sub>2</sub> SiO <sub>4</sub>	0.00	Fs:	3.00		$\mathrm{Ca}_2\mathrm{Si}_2\mathrm{O}_6$

spinel octahedra and adjacent olivine grains. A pressure estimate is rather problematic because of distinct mineral disequilibria caused by strongly different diffusion coefficients for the elements of concern in different minerals (cf. Franz et al. 2000). An intersection of the recent geothermal gradient of the Bismarck Archipelago with the  $\rm K_D$ -line of the T BKN thermometry results in a pressure–temperature (P–T) estimate of 820 °C at 16.6 kbar.

Sample 56-2X is a coarse-equant spinel lherzolite, which is crosscut by a 5-mm-wide metasomatic vein consisting of phlogopite, hornblende, secondary clinopyroxene, garnet, anorthite, and mackinawite. An explicit description of the mineral chemistry and the metasomatic reactions, which generated this vein, is presented elsewhere (Franz et al. 2000). The primary mineral assemblage mainly consists of coarse-grained olivine ( $Fo_{91-92}$ ), which again shows the typical, retrograde zoning of Ca (100 ppm Ca in the core vs 34 ppm Ca in the rim). Ortho- and clinopyroxene do not show exsolutions under the petrographic microscope but display distinct chemical zoning. Orthopyroxene ( $X_{Mg}0.91$ – 0.92) shows a decrease of the wollastonite component from core to rim (1.2 mol% vs. 0.47 mol% Wo) as well as decreasing Al and Cr concentration in the same direction. Clinopyroxene displays a composition of Wo<sub>46</sub>En<sub>52</sub>Fs<sub>2</sub> in the core and Wo<sub>43</sub>En<sub>54</sub>Fs<sub>3</sub> in the rim section. These features again testify to distinct cooling processes in the mantle. Intergranular spinel octahedra display typical zonation patterns with abruptly decreasing Cr and Al contents at the expense of Fe<sup>3+</sup>, which is especially prominent near the metasomatic vein. Spinel needles are present in some of the large olivine grains remote from the metasomatic vein. Selected mineral analyses are presented in Tables 2 and 3 as well as in Franz et al. (2000).

Thermobarometry applied to core analyses of pyroxenes yields a temperature of ~930 °C (at 10 kbar) using the T BKN calibration, whereas analyses from the corresponding rim sections indicate cooling processes (720 °C at 10 kbar). Temperatures calculated with the olivine-spinel geothermometer of Ballhaus et al. (1991) are in the range of 750 °C for the core sections of the minerals, whereas analyses from the rim sections yield 720-740 °C. The P-T estimate for the metasomatic overprint is 720–770 °C at 4–8 kbar (Franz et al. 2000). Quite remarkable is the elevated oxygen fugacity  $(\Delta \log f_{O_2} = 1.3 \text{ to } 2.5; \text{ cf. Franz et al. } 2000) \text{ during}$ the metasomatic overprint, which is a typical feature of the metasomatic peridotites of this region resulting from he hydrous fluids generated by dehydration of the underlying subducted slab (see also McInnes et al. 2000).

### Inclusions in olivine

Both samples reveal numerous inclusions in olivine, which commonly have sizes of less than a micron. TEM

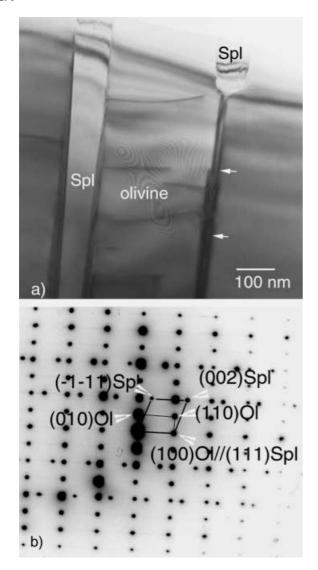
investigations show that the majority of these inclusions are different types of spinel.

- 1. The most common are spinel needles, which display a crystallographic orientation in the olivine grains. The orientation relationship is given by:  $[001]_{Ol}//[1\overline{10}]_{Spl}$ and  $(100)_{Ol}/(111)_{Spl}$  (see Fig. 3). The needles are elongated in a parallel [010] direction in olivine and oriented parallel to the (111) plane of spinel. The precipitates are semi-coherent because only the d-spacings of the parallel  $(111)_{Spl}$  $(100)_{O1}$ (d = 0.4762 nm)and (d = 0.481 nm) have a similar lattice spacing. The needles have a typical length of 8 µm (see Fig. 2) although some olivine grains bear irregularly shaped spinel needles with serrated rims and a length exceeding 25 µm. TEM investigations reveal a lenticular shape of the needles with widths and depths of 50–200 nm. Because of their small size, the composition of the spinel needles can only be determined by AEM. In sample 56-2P, spinel needles yield rather uniform compositions with  $X_{Mg}$  values of 0.61–0.72 and  $X_{Cr}$  values of 0.49–0.55, thus resembling the intergranular spinel octahedra (Table 1). Differences in  $X_{Mg}$  arise from secondary fluorescence effects of the hosting olivine leading to elevated SiO<sub>2</sub> and consequently increased MgO-concentrations because the chemical composition is normalized to 100%. Consequently, because of these analytical limitations, an estimate of the Fe<sup>3+</sup>-content of the spinel needle is affected by an error.
- 2. In sample 56-2X, spinel needles show a wide variability in composition ranging from specimens with intermediate Cr and Al contents (~39 wt% Cr<sub>2</sub>O<sub>3</sub> and 21 wt% Al<sub>2</sub>O<sub>3</sub>) to Cr-rich types (48.5–55.8 wt% Cr<sub>2</sub>O<sub>3</sub> and 7.2–16.4 wt% Al<sub>2</sub>O<sub>3</sub>). As evident from the chemical analyses (Table 2 and Fig. 7), the spinel needles with intermediate Cr- and Al-concentrations resemble the intergranular spinel octahedra.
- 3. Small spinel octahedra included in olivine with typical diameters of 200–400 nm occur in both samples and reveal the same orientation relationship with respect to olivine as the spinel needles (Fig. 4). They are distinguished from the spinel needles by their different chemical composition with an extreme enrichment in Al (41.2–59.8 wt% Al<sub>2</sub>O<sub>3</sub>) and low Cr concentrations (2.0–5.3 wt% Cr<sub>2</sub>O<sub>3</sub>; Tables 1 and 2). As evident from the SiO<sub>2</sub> contents of these grains, which are in the range of 2.2 and 5.3 wt%, a secondary fluorescence of the hosting olivine cannot be excluded.
- 4. Tiny idiomorphic magnetite grains, which appear in sample 56-2X, show diameters of  $\sim$ 200 nm and exhibit the same orientation relationship with the host olivine as that of spinel needles:  $[001]_{Ol}/[1\overline{10}]_{Spl}$  and  $(100)_{Ol}/(111)_{Spl}$  (Fig. 5). This orientation relationship was earlier reported for magnetite inclusions in olivine by Champness (1970). The magnetite grains have a certain amount of Cr and Mg (Table 2) and are often associated with cavities. In one case NaCl was found in such a cavity together with orthopyroxene. The same mineral paragenesis of magnetite, orthopyroxene and NaCl in olivine was described by Puga et al. (1999).

iable 2 Representative analyses of spirier from sample nicroprobe analysis; n number of analyses. T BBG and 56-2X 56-2X 56-2X	iber of ar	nicroprobe analysis; $n$ number of analyses. T BBG and Da $56-2X$ $56-2X$ $56-2X$ $56-2X$ $56-2X$	1)	56-2X	56-2X	56-2X	56-2X	56-2X	6-2X 56-2X	56-2X	56-2X	56-2X	56-2X
Al-Spl N Core C	, ~ 0 ,	Magnetite Core	Lamella	Lamella	Lamella	Lamella	Lamella 5	Spl matrix Rim	Spl Matrix Rim	Spl matrix Rim	Spl matrix Rim	Spl matrix Core	Spl matrix Rim
		AEM	AEM	AEM	AEM	AEM	AEM	AEM	AEM	AEM	AEM	EMP	EMP
4.30		2.45	2.00	1.10	1.50	2.00	1.40	0.40	0.90	0.40	0.50	0.14	0.48
0.00		1.51	0.00	0.00	0.80	0.00	0.00	0.00	0.00	0.00	0.00	0.15	0.63
41.20		5.38	12.80	5.90	5.40	12.80	22.10	21.60	18.50	18.90	19.80	27.99	20.80
4.10		3.11	46.70	59.10	57.80	46.70	42.90	42.90	39.50	51.20	49.50	40.46	36.69
16.32		56.85	6.07	5.88	4.75	6.07	4.32	5.55	11.84	0.00	0.00	0.64	10.52
15.40		11.13	12.30	11.20	10.30	12.30	14.60	12.20	12.20	9.70	10.00	12.32	11.66
3.30		0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01
0.00		0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
16.91		19.58	18.03	17.41	19.93	18.03	15.12	17.20	17.44	19.70	20.10	17.63	17.87
0.00		0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	90.0	0.14
0.00		0.00	0.00	0.00	0.00	00.00	0.00	0.00	0.00	0.00	0.00	0.09	0.25
01.53		100.00	100.90	100.59	100.47	100.91	100.43	99.85	100.38	06.66	06.66	99.49	99.04
0.118		0.083	0.064	0.036	0.050	0.064	0.042	0.012	0.028	0.013	0.016	0.004	0.015
0.000		0.039	0.000	0.000	0.020	0.000	0.000	0.000	0.000	0.000	0.000	0.003	0.015
		0.710	0.480	0.231	0.213	0.480	1.039	1.051	0.084	0.708	1 238	0.000	0.7/4
		1.456	0.217	0.147	0.119	0.217	0.000	0.130	0.280	0.000	0000	0.075	0.250
		0.565	0.584	0.554	0.513	0.584	0.659	0.565	0.571	0.460	0.472	0.557	0.549
		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
0.000		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
0.389		0.557	0.480	0.483	0.557	0.480	0.383	0.447	0.458	0.524	0.532	0.447	0.472
0.000		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.003
0.000		0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.002	9000
3.000		3.000	$\frac{3.000}{2.0}$	3.000	$\frac{3.000}{\hat{i}}$	3.000	3.000	$\frac{3.000}{2.0}$	$\frac{3.000}{2.2}$	2.990	2.996	$\frac{3.000}{2.0}$	$\frac{3.000}{2.000}$
0.62		0.50	0.55	0.53	0.48	0.55	0.63	0.56	0.55	0.47	0.47	0.55	0.54
0.00		0.20	10.01	0.8/	0.00	1001	0.3/	0.3/	10.0	10.0	0.02	0.492	10.34
523 10	$\equiv$	001	753	823	755	754	752	680	729	595	583	590	687
		5.69	2.21	1.41	1.17	2.21	0.88	1.43	2.71	ı	ı	-2.10	2.64

Table 3 Representative analyses of olivine, pyroxene, melt inclusions, and a cavity filling from sample 56-2X. Fe<sup>3+</sup> estimated according to the method of Droop (1987). AEM analytical electron microscopy; EMP electron microprobe analysis; n number of analyses. T BBG and Delta log fo<sub>2</sub> are Ol–Spl thermometry and Ol–Spl–Opx oxygen barometry of Ballhaus et al. (1991)

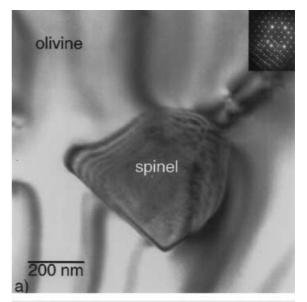
Dalliaus et al. (1991)	(16)												
(%1%)	56-2X OI 1 avg. $(n = 93)$ EMP	Cpx (wt%)	56-2X Cpx I Core EMP	Opx (wt%)	56-2X Opx I Core EMP	56-2X Opx II Core AEM	Melt inclusions in olivine (wt%)	56-2X Melt Gl Core AEM	56-2X Melt G4 Core AEM	56-2X Melt G5 Core AEM	56-2X Melt A3 Core AEM	Cavity filling: NaCl+Ol+Opx (wt%)	56-2X Center AEM
SiO <sub>2</sub> TiO <sub>2</sub> Al <sub>2</sub> O <sub>3</sub> Cr <sub>2</sub> O <sub>3</sub> MgO CaO MnO FeO NiO Total Cations (O = 4) Si Ti Al Cr Ni Ni Ti Al Cr Mg Cr Ni Cr Mg Cr	40.16 0.00 0.00 0.00 0.02 0.02 0.03 0.34 98.51 0.989 0.0000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.0	$\begin{array}{llllllllllllllllllllllllllllllllllll$	54.23 0.23 2.15 0.00 17.64 22.98 0.07 1.79 0.36 0.02 0.038 0 = 6) 1.954 0.006 0.091 0.006 0.091 0.006 0.094 0.026 0.094 0.026 0.026 0.027 0.025 0.002 0.025 0	SiO <sub>2</sub> TiO <sub>2</sub> Al <sub>2</sub> O <sub>3</sub> Cr <sub>2</sub> O <sub>3</sub> Fe <sub>2</sub> O <sub>3</sub> Fe <sub>2</sub> O <sub>3</sub> MgO CaO Na <sub>2</sub> O K <sub>2</sub> O Tiotal Total Ti Al Fe <sup>2</sup> + Ca Mg Ca Mn Mg Ca Ca Mg Ca Ca Mg Ca Ca Ca Mg Ca	57.55 0.09 1.46 0.34 0.00 35.27 0.63 0.14 5.20 0.00 0.00 0.00	55.55 0.00 0.00 8.00 37.30 0.00 0.00 0.00 100.85 1.897 0.000	SiO <sub>2</sub> $+2.90$ $+7.10$ $28.20$ $+0.80$ $Al_2O_3$ $1.90$ $1.60$ $1.90$ $1.70$ $Cr_2O_3$ $2.60$ $1.60$ $3.50$ $4.50$ $MgO$ $2.60$ $36.60$ $2.60$ $30.60$ $2.60$ $30.60$ $2.60$ $30.60$ $2.60$ $30.60$ $2.60$ $3.10$	42.90 1.90 2.60 35.40 0.90 1.30 99.90 exchange the 10.0 10.0	47.10 1.60 30.60 1.70 1.430 1.80 0.00 1.30 100.00 ermometry a 10.0 1189	28.20 1.90 3.50 26.00 0.80 34.30 2.30 0.00 2.40 99.40 10.0 11.0 1699	40.80 1.70 4.50 30.60 3.10 1.30 0.00 2.60 100.10 1205 1149	SiO <sub>2</sub> AI <sub>2</sub> O <sub>3</sub> Cr <sub>2</sub> O <sub>3</sub> MgO CaO ReO Na <sub>2</sub> O K <sub>2</sub> O Cl <sub>2</sub> O Total	47.90 0.00 0.00 0.00 0.00 6.20 3.10 10.70 100.10

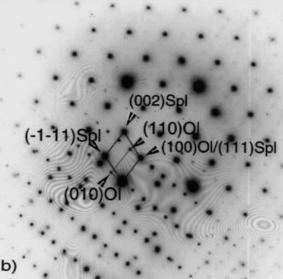


**Fig. 3** a TEM bright-field image showing two spinel needles in olivine (sample 56-2X). Note that the spinel needles run parallel to the low-angle grain boundary in olivine. The interface is oriented parallel to the beam, therefore no dislocation lines are visible. **b** Corresponding  $[001]_{Ol}$  diffraction pattern revealing the orientation relationship between spinel and olivine  $(100)_{Ol}//(111)_{Spl}$ , and  $[001]_{Ol}//[1\overline{10}]_{Spl}$ 

The secondary orthopyroxene in sample 56-2X reveals a rather peculiar composition with Ca, Na and Al contents below the detection limit of the AEM, and charge balance constraints point to a mere presence of ferric iron (cf. Table 3). The analysis of NaCl from the cavity is characterized by strong secondary fluorescence and/or influence of the underlying olivine grain (Table 3).

Beside orthopyroxene and NaCl, cavities in olivine often contain idiomorphic calcite and CaCl<sub>2</sub>. Furthermore, fluid inclusions are present along healed cracks linked with dislocations. The olivine crystals show a remarkable microstructure. They exhibit long, straight dislocation lines, which indicates deformation by dislocation glide. Nearly all of the olivine grains are subdi-

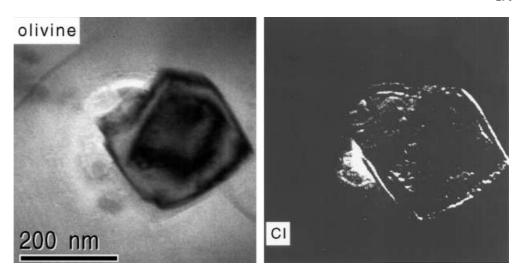


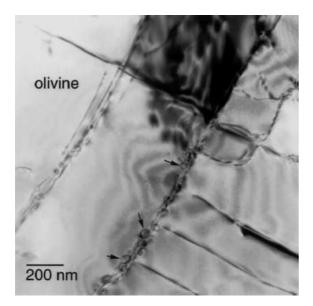


**Fig. 4 a** TEM bright field image showing an Al-spinel octahedron in olivine of sample 56-2P with diffraction pattern inserted **b** [001]<sub>OI</sub> diffraction pattern revealing the orientation relationship between spinel and olivine

vided into subgrains by parallel low-angle grain boundaries (Fig. 6). The subgrains have a typical width of  $\sim$ 1 µm. Very often, fluid inclusion bubbles or melt inclusions are associated with the low-angle grain boundaries. The melt inclusions have an ultramafic composition with variable concentrations of SiO<sub>2</sub> (28–47 wt%; Table 3). According to the Fe–Mg-exchange geothermometer of Ford et al. (1983), temperatures of  $\sim$ 1100–1300 °C (TMg, Table 3) were calculated for these melt inclusions and the surrounding olivine, which points to the generation of that melt by the host basalt. Healed cracks are decorated with fluid inclusion bubbles. Furthermore, the spinel/olivine interface shows numerous fluid inclusion bubbles, which are commonly linked with dislocations.

Fig. 5 Elemental map of a magnetite inclusion in olivine of sample 56-2X using Cl. In the *upper left corner* is the corresponding energy-filtered bright field image (zero-loss filtering, energy window 10 eV). Note the chlorine in the cavity, which stems from NaCl





**Fig. 6** TEM dark-field image of two parallel low-angle grain boundaries in olivine. Note the tiny fluid inclusion bubbles decorating the low-angle grain boundary

# **Discussion**

The literature contains several models that explain the formation of spinel needles and symplectites. The following section gives an evaluation of these models with respect to the formation of the observed spinel inclusions in the xenoliths of TUBAF seamount.

- 1. Breakdown of garnet inclusions in olivine, as postulated by Bell et al. (1975), cannot explain the formation of any spinel inclusions in olivine. Such a breakdown usually generates symplectites of olivine, spinel as well as ortho- and clinopyroxene, which were not observed within olivine in the TUBAF xenoliths.
- 2. Breakdown of phlogopite might be responsible for the formation of the aluminous spinel octahedra in both investigated samples. According to Pasteris (1983), the reaction

$$\begin{split} 4KMg_3\big[(OH)_2AlSi_3O_{10}\big] &\Longleftrightarrow 5Mg_2[SiO_4] + 2MgAl_2O_4\\ phlogopite & forsterite & spinel\\ &+ (2K_2O + 4H_2O + 7SiO_2) \end{split}$$

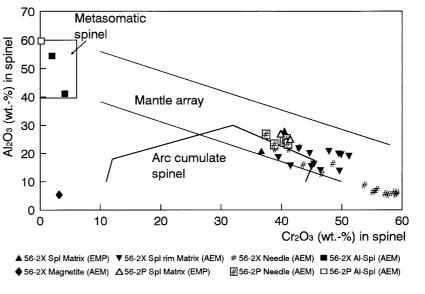
which he observed in the DeBeers kimberlite, may lead to the generation of spinel similar in composition to the aluminous spinel octahedra in the TUBAF samples. The exact time of this event, however, cannot be determined. Phlogopite breakdown could have occurred during the plume-induced heating of the asthenosphere, during the initial formation of the mid-ocean ridge or during the influence of fluids emerging from the Quaternary host basalt. As the minimum temperatures for the breakdown of phlogopite are ~850 °C (Wagner and Velde 1987) the metasomatism by the slab-derived aqueous fluids can surely be excluded for the formation of the aluminous spinel. Another possibility is the influence of metasomatic fluids, which may have produced this spinel by complex cation exchanges. Metasomatic aluminous spinel with a similar composition was described in kimberlites by Haggerty (1988; Fig. 7).

3. Oxidation and cellular decomposition (Putnis 1979) can be rejected for the generation of the chromian and the aluminous spinel because the shape of the inclusions does not resemble at all the products of such a process (see Putnis 1979; Moseley 1984) and furthermore, neither the composition of these spinel inclusions (which should be magnetite in case of the oxidation) nor accompanying pyroxene was observed in the TUBAF xenoliths. However, the few magnetite grains may well be generated by such an oxidation. The finding of excavations with NaCl phase near the magnetite and the presence of associated orthopyroxene points to a reaction:

$$6(Fe, Mg)_2[SiO_4] + O_2 \iff 2Fe_3O_4 + 3Mg_2[Si_2O_6]$$
  
olivine oxygen magnetite enstatite

with a hydrous, saline fluid phase acting as the oxidizing agent (for the reaction mechanism see Champness 1970). Puga et al. (1999) describe magnetite inclusions in olivine from ophiolitic metagabbros associated with

**Fig. 7** Al<sub>2</sub>O<sub>3</sub> vs Cr<sub>2</sub>O<sub>3</sub> diagram for spinel of samples 56-2P and 56-2X. Fields of arc cumulate spinel, metasomatic spinel and mantle array after Conrad and Kay (1984), Haggerty (1988), and Kepezhinskas et al. (1995)



orthopyroxene and NaCl. There, NaCl is indicative for seawater influence on the formation of magnetite in olivine. A coupled exsolution process of fluid and spinel in olivine as proposed by Green (1985; see also Burns 1975) seems rather improbable for the TUBAF xenoliths. According to these authors, the existence of dissolved carbon and O<sup>-</sup> in olivine may lead to the formation of spinel inclusions during decompression. However, such a mechanism would always be connected with the formation of syngenetic CO<sub>2</sub> fluid inclusions and in part with the generation of native carbon, which both have not been observed near the magnetite octahedra nor the other spinel inclusions in olivine of the TUBAF xenoliths.

- 5. Another possibility to explain the presence of magnetite inclusions in olivine would be the decomposition of high-P, Fe<sup>3+</sup>-bearing olivine as observed by Zhang et al. (1981, 1999) in ultrahigh-pressure rocks. For the TUBAF xenoliths, this mechanism seems improbable as there are no indications of any high-pressure event. The same applies to the generation of spinel needles by exsolution of wadsleyite solid solution during decompression (Zhang et al. 1999).
- 6. The generation of chromian spinel inclusions in olivine was investigated by Duke (1976), who found that olivine crystallizing from a chromium rich melt may incorporate elevated amounts of Cr<sup>3+</sup>. During subsequent annealing chromian spinel may exsolve (Arai 1978). However, this model cannot be applied to the TUBAF xenoliths because they are not magmatic cumulates, but represent depleted lithospheric mantle, which equilibrated in the stability field of spinel peridotite. For the same reason, a simultaneous crystallization of chromian spinel and olivine from a mafic parental magma can be excluded.
- 7. A process that may lead to the generation of chromian spinel inclusions in olivine could result from cooling and decompression of Cr- and Al-rich olivine in the mantle under the mid-ocean ridge. At high temper-

ature and pressure, olivine may incorporate elevated amounts of Cr and Al, as shown by Dodd (1973) for olivine of the Sharps chondrite. Based on analytical data, Dodd (1973) proposed a coupled substitution of Al +  $\text{Cr}^{3+} \iff \text{Si}^{4+} + (\text{Mg}, \text{Fe}^{2+})$  in olivine. On cooling, unmixing processes could have led to the formation of the Cr- and Al-rich spinel needles found in the olivine of the TUBAF xenoliths via the reaction:

This mechanism explains the presence of single phase spinel inclusions as well as their specific chemical composition. A quantification of the solubility of Al and Cr in olivine at elevated pressures and temperatures has been undertaken by Köhler (1989), who showed that olivine may incorporate up to 710 ppm Cr<sub>2</sub>O<sub>3</sub> and 450 ppm Al<sub>2</sub>O<sub>3</sub> at 1200 °C and 20 kbar. These P–T values would be similar to the conditions in the upper oceanic asthenosphere during the formation of the basaltic magma in the course of mid-ocean ridge spreading. On cooling and decompression, solubility for Al and Cr is decreasing (see Köhler 1989), which may have given way to the exsolution of the spinel needles.

The question is whether these low contents of Cr and Al in olivine were really sufficient to produce the observed spinel needles by exsolution. For this reason, the Al- and Cr-contents of selected olivine grains were determined by microprobe analyses with elevated beam current acceleration voltages and counting times (see above) along traverses through selected olivine grains. For the olivine of sample 56-P, an average content of 15 ppm for Cr<sub>2</sub>O<sub>3</sub> and of 20 ppm for Al<sub>2</sub>O<sub>3</sub> were measured and similarly low contents of 20 ppm for Cr<sub>2</sub>O<sub>3</sub> and Al<sub>2</sub>O<sub>3</sub> were detected in olivine of sample 56-2X (see Tables 1 and 3). This is close to the detection limit of the

probe and shows that olivine from both samples was almost devoid of these elements. To determine the amount of Cr and Al in the spinel needles, all needles from an inclusion-rich olivine grain from sample 56-2P were counted and their lengths were measured under the optical microscope. The procedure revealed a number of 1415 needles with an average length of  $\sim 8 \mu m$ . As indicated by the TEM studies, the needle-shaped spinel crystals have a maximum width and depth of 200 nm. Using these data, the total volume of all needles was calculated. Because of their tiny width and depth, the spinel needles occupied a volume of only 0.0013% in the hosting olivine. Even in sections of the olivine grains with an accumulation of spinel inclusions, their volume did not exceed 0.0044%. As evident from the AEM analyses, a typical chromian spinel needle yields an average of 25 wt% Al<sub>2</sub>O<sub>3</sub> and 40% wt% Cr<sub>2</sub>O<sub>3</sub>, and thus, all observed needles cause an increase of about 3 ppm Al<sub>2</sub>O<sub>3</sub> and 5 ppm Cr<sub>2</sub>O<sub>3</sub> in the hosting olivine grain. Even in sections with an accumulation of spinel inclusions, the increase does not exceed 11 ppm Al<sub>2</sub>O<sub>3</sub> and 18 ppm Cr<sub>2</sub>O<sub>3</sub> and therefore, these needles may well have formed by exsolution. Any additional amount of Cr and Al contained in the high-temperature olivine could have left the grain by diffusion during cooling forming the large, intergranular spinel octahedra.

These interpretations are supported by the Al<sub>2</sub>O<sub>3</sub> vs Cr<sub>2</sub>O<sub>3</sub> diagram for spinel (Fig. 7), which reveals the variability in composition of the different spinel types and shows that all chromian spinel (i.e. intergranular spinel and spinel needles) plot in the mantle array outlined by Kepezhinskas et al. (1995). In contrast to this, aluminous spinel plots in the field of metasomatic spinel, which, however, also includes spinel generated by breakdown of aluminous silicates (see Wagner and Velde 1987; Haggerty 1988; Mukhopadhyay 1991).

#### **Conclusion**

The presence of numerous spinel inclusions in olivine from mantle xenoliths of the TUBAF seamount can be assigned to genetically different processes. Magnetite octahedra, which are usually accompanied by orthopyroxene and formerly fluid-filled cavities are supposed to have formed by oxidation of the olivine grain. The oxidizing agent may have been generated by dehydration of the underlying, subducted Pacific plate or may be derived from the volatile rich, Quaternary volcanism (McInnes et al. 2000), which transported the xenoliths to the surface.

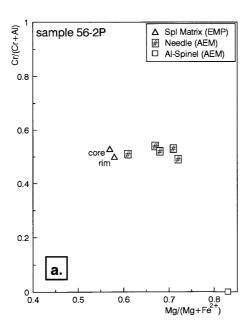
Rare aluminous spinel octahedra are probably the breakdown product of an Al-rich phase such as phlogopite or may also have developed under the influence of a metasomatic fluid. However, because of missing textural relationships and the absence of accompanying critical phases, the time of their generation cannot be determined.

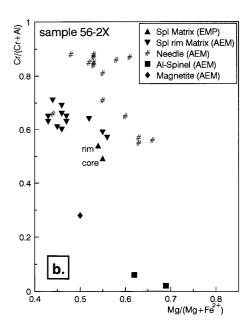
The majority of the spinel inclusions, consisting of chromian spinel needles, are assumed to have formed by exsolution processes. The mantle peridotite xenoliths

from TUBAF seamount in the Bismarck Archipelago of Papua New Guinea give evidence for distinct cooling processes, which are interpreted to have taken place during the formation of the mantle lithosphere under the mid-ocean ridge (Franz et al. 2000). The formation of lithospheric mantle is well described by models of Houseman and McKenzie (1982), Houseman (1983a, b), or Rabinowicz et al. (1984), who postulate a rolling mill effect of the mantle at mid-ocean ridges. According to these authors partial melting and melt extraction processes affect the ultramafics during their uplift, thus creating the depleted lithospheric mantle. As evident from investigations of meteorites (Dodd 1973) and from experiments of Köhler (1989), olivine may incorporate elevated concentrations of Cr and Al at higher temperatures. During uprise from the hot asthenosphere, olivine must have exsolved the observed Cr- and Al-rich spinel needles. An argument for such exsolution processes is the crystallographic orientation of the needles in the olivine grains. Volumetric calculations have shown, that the amount of Cr and Al of high-temperature olivine is sufficient to have produced these spinel exsolutions. During the thermal equilibration in the upper mantle lithosphere, the spinel needles, as well as the large intergranular spinel octahedra, experienced a resetting of their chemistry, which is visible in sample 56-2P. Spinel needles reveal a very similar composition to the intergranular spinel specimen and deviations mainly result from analytical inaccuracy (i.e. incorporation of the hosting olivine in the analysis). Because of this analytical error, a calculation of Fe–Mg-exchange thermometry between the spinel needles and the olivine results in a wide scatter of the temperatures (i.e. 713– 971 °C; see Table 1).

Although spinel needles in sample 56-2P reveal about the same X<sub>Cr</sub>-values as the large intergranular octahedra, needles in sample 56-2X in part show significantly higher X<sub>Cr</sub>-values than the large grains in the matrix (Fig. 8a, b). This variability in X<sub>Cr</sub> can probably be ascribed to the metasomatism of fluids generated by dehydration of the underlying slab, which also caused the veining of this sample. Similar patterns of Cr-enrichment are revealed by microprobe analyses from the rim sections of intergranular spinel octahedra, which is especially prominent near the metasomatic vein of this sample (Fig. 8b). Compared with the Cr-rich needles, X<sub>Cr</sub> of the intergranular octahedra still may seem rather low, but this is just a question of the analytical resolution. The microprobe beam never reaches the outermost rim section of the grain and only the use of AEM gives an insight into these domains. According to these AEM analyses, the composition of the outermost rim of the intergranular octahedra shows great resemblances to the composition of the tiny spinel needles, which highlights late processes of equilibration (see Table 2). Despite the analytical error of the AEM and despite the chemical re-equilibration of the spinel needles, most Fe-Mg-exchange temperatures with the hosting olivine are in the range of 700–800 °C

Fig. 8a, b  $X_{Cr}$  vs  $X_{Mg}$  diagrams for spinel of samples a 56-2P and b 56-2X





(see Table 2), thus corresponding to the temperatures estimated for the metasomatic event (see above and Franz et al. 2000). Furthermore, an elevated oxygen fugacity is revealed by these spinel needles applying the barometry of Ballhaus et al. (1991), which is also indicative of the metasomatism (see Table 2 and cf. Franz et al. 2000; McInnes et al. 2000).

These observations show that the formation of spinel inclusions and their compositional modification may be caused by different processes, which can eventually be observed in the same sample. Furthermore, it becomes evident that a knowledge of the geological setting and the physico-chemical processes affecting the rocks can be the key for the understanding of all the petrological observations.

Acknowledgements The authors thank Karin Paech (Potsdam) for her extraordinary skill in TEM specimen preparation and Klaus-Peter Becker (Freiberg) for the assistance with the microprobe. The manuscript was considerably improved by the inspiring reviews of Pamela Champness and Wolfgang Müller. We also want to thank Gerhard Brey and Thomas Stachel (Frankfurt) for helpful remarks and fruitful discussion. We owe great thanks for the logistic and scientific support to the master, officers and crew of research vessel RV *Sonne* during our research activities in the New Ireland basin. Special thanks are due to Peter Herzig (Freiberg), chief scientist of the research campaign, who enabled L.F. to participate in the Edison II cruise. Principal funding for this project was provided by the German Federal Ministry for Education and Science (BMBF Grant 03G0133A to Peter Herzig).

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