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Clinopyroxenes from metasomatized spinel-peridotite mantle xenoliths from Nemby (Paraguay): crystal chemistry and petrological implications

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Summary

The Nemby mantle xenoliths are represented by spinel peridotites variably affected by "metasomatic processes" in Proterozoic times (1.2–0.5 Ga). Clinopyroxene compositional variations mainly reflect variably degrees of basalt extraction and mantle "metasomatic processes". Clinopyroxene crystal chemistry allows to estimate the equilibration pressure by V(Cell) and V(M1) relationship. The investigated clinopyroxenes yielded a pressure range of 12–18 kb. Therefore, the Nemby xenoliths are representative of about 20 km thickness of subcontinental lithospheric mantle of the South America platform at Tertiary times (ca. 46 Ma).

Zusammenfassung

Klinopyroxene aus metasomatischen Spinell-Peridotit-Mantel Xenolithen aus Nemby (Paraguay): Kristallchemie und petrologische Bedeutung

Die Mantelxenolithe aus Nemby sind Spinellperidodite, die in verschiedenem Ausmaß im Proterozoikum (1.2 bis 0.5 Ga) durch "metasomatische Prozesse" beeinflusst wurden. Variationen in der Zusammensetzung der Klinopyroxene lassen variable Ausmaße von Basaltextraktion und Mantelmetasomatose erkennen. Die Kristallchemie der Pyroxene ermöglicht es, den Gleichgewichtsdruck durch V(Cell) und V(M1) Beziehungen zu bestimmen. Die untersuchten Klinopyroxene zeigten Drucke im

Bereich von 12 bis 18 kb. Die Nemby Xenolithe sind deshalb repräsentativ für einen ungefähr 20 km dicken subkontinentalen lithosphärischen Mantel der südamerikanischen Plattform im Tertiär (ca. 46 Ma).

Introduction and petrological notes

Mantle spinel-peridotite xenoliths occur in the Tertiary (46 Ma) melanephelinite plug from Nemby (southeast of Asuncion, Paraguay; *Comin-Chiaramonti* et al., 1991). These xenoliths range from lherzolite (modal clinopyroxene 5–10 vol%) to harzburgite (clinopyroxene less than 2 vol%), and show protogranular texture and the typical four-phases assemblage: olivine (ol), orthopyroxene (opx), clinopyroxene (cpx) and spinel (sp) in order of decreasing amount (Table 1). Cpx shows variable morphology, ranging from well preserved to relict crystals frequently with spongy rims. Occasionally, cpx shows exsolution lamellae of sp or more rarely of opx, and sometimes contain glassy drops. In addition to these features, some studied xenoliths contain glassy microcrystalline rounded blebs and/or interstitial brown or colorless glass. The blebs, developed around lobate spinels, are constituted of ol, cpx, euhedral Cr-sp microlites set in a glassy-feldspathic matrix.

According to *Demarchi* et al. (1988), the Nemby mantle xenoliths were distinguished into two suites, characterized by low and high potassium and incompatible element (IE, ppm) content (LK and HK suites, respectively). Generally, the IE enrichment (i.e. LK: K = 166–1660, Rb = 0.5–8, Sr = 7–20, Ba = 11–20, Nd = 0.04–0.30; HK: K = 1661–4898, Rb = 2–12, Sr = 14–75, Ba = 18–65, Nd = 0.56–2.42; *Petrini* et al., 1994; *Comin-Chiaramonti*, unpublished data) is higher than those of amphibole-bearing spinel peridotite xenoliths from Nunivak (*Roden* et al., 1984). LK and HK Nemby xenolith suites are also characterized by low and high content of glassy blebs and/or glassy drops in clinopyroxenes, respectively (*Comin-Chiaramonti* et al., 1986). The latter Authors interpreted the glassy blebs as due to the melting of hydrous phases (amphibole and/or phlogopite) during the mantle xenolith ascent to the surface.

Petrini et al. (1994) documented that the chemical equilibrium among opx, cpx, ol and sp, in most of the Nemby xenoliths, was almost complete, and that the last equilibration occurred in a relatively small temperature range, i.e. 1000 ± 50 °C.

Table 1. *Modal analyses of Nemby xenoliths;* ol *olivine,* opx *orthopyroxene,* cpx *clinopyroxene,* sp *spinel;* Fo% *forsterite content*

Sample	ol	opx	cpx	sp	ol Fo%	opx $Mg/(Mg+Fe_t)$	cpx $Mg/(Mg+Fe^{2+})$	sp $Cr/(Cr+Al)$
LK3192	70.3	17.5	9.8	2.4	90.5	0.912	0.910	0.126
LK3221	65.2	23.9	7.1	3.8	90.5	0.912	0.942	0.215
LK3227	77.2	18.1	3.2	1.5	91.5	0.921	0.958	0.271
LK3269	76.4	21.1	1.9	0.6	91.7	0.921	0.959	0.307
HK3311	67.7	20.5	11.0	0.8	89.8	0.903	0.904	0.183
HK3307	76.9	14.5	6.6	2.0	90.6	0.916	0.931	0.214
HK3211	75.5	19.9	3.4	1.2	91.0	0.919	0.956	0.400
HK3284	84.1	12.5	1.0	2.4	87.9	0.901	0.968	0.704

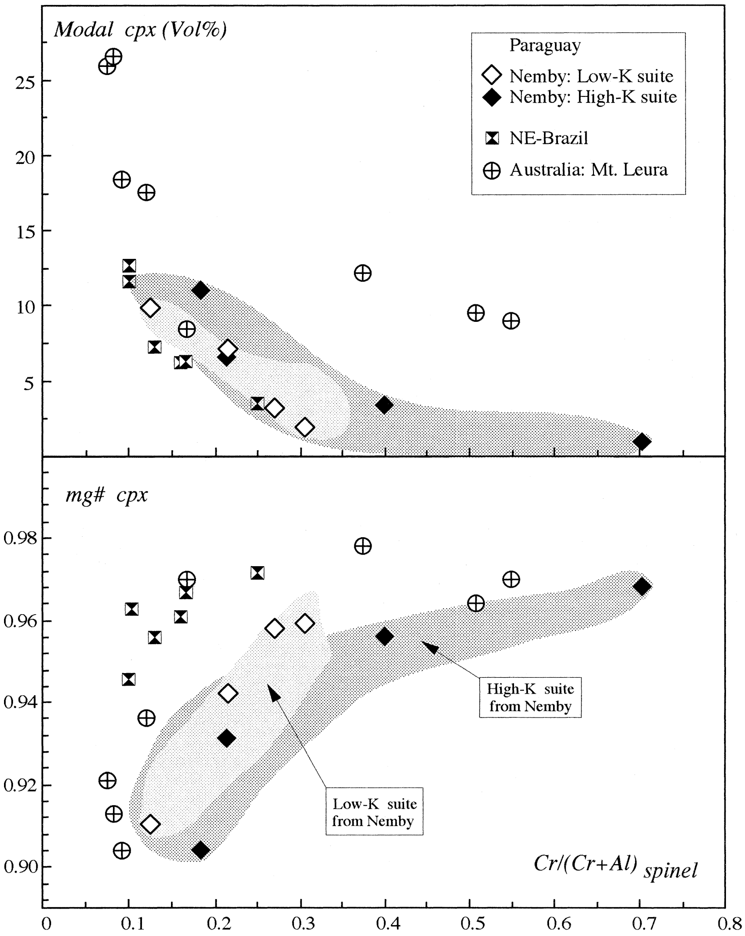

Fig. 1. Cr/(Cr + Al) of spinels vs. mg# value of cpx and vs. cpx modal content. Mt. Leura suite (*Dal Negro* et al., 1984), NE-Brazil suite (*Princivalle* et al., 1989) are shown for comparison

An exception is probably represented by the clinopyroxene 3284 (HK suite) which closely approaches a diopsidic composition.

The geochemical characteristics of LK and HK cpx from Nemby xenoliths were interpreted as the result of "metasomatic processes" which variably affected the mantle in Proterozoic times (1.2–0.5 Ga; *Comin-Chiaramonti* et al., 1986; *Demarchi* et al., 1988; *Petrini* et al., 1994; *Rivalenti* et al., 1996). The metasomatic imprint involved mantle peridotites variably depleted in basalt component.

In Fig. 1, which includes the entire fields of LK and HK xenoliths (*Demarchi* et al., 1988), the variation of the Cr/(Cr + Al) of spinels is plotted vs. the cpx mg# ratio [mg#:Mg/(Mg + Fe^{2+})] and the modal cpx content, respectively (cf. Table 1). In Fig. 1 the Nemby cpx are compared with those from spinel peridotite xenoliths of Mt. Leura (Victoria, Australia; *Dal Negro* et al., 1984) and NE-Brazil (*Princivalle* et al., 1989). The Mt. Leura and NE-Brazil cpx suites belong to mantle xenoliths which suffered variable degree of incompatible-element enrichment, quite lower than that of the LK and HK Nemby xenolith suites (*Rivalenti* et al.,

Table 2. *Cell parameters and site geometry of the LK and HK cpx. N. Obs. Rifl. number of observed reflections; R. Fin.*$=\|\Sigma Fo| - |\Sigma Fc\|/|\Sigma Fo|$ *%;* σ_θ^2 *tet. and* σ_θ^2 *oct.: variance of the tetrahedral and octahedral angles* (Robinson *et al., 1971), respectively;* $\Delta M2 = (M2\text{-}O3,C2) - [(M2\text{-}O3,C1) + (M2\text{-}O1) + (M2\text{-}O2)]/3$*. Errors in parentheses refer to last digits*

Sample	LK3192	LK3221	LK3227	LK3269	HK3311	HK3307	HK3211	HK3284
a_o (Å)	9.691 (4)	9.712 (3)	9.723 (2)	9.715 (2)	9.698 (3)	9.719 (3)	9.714 (2)	9.729 (3)
b_o (Å)	8.864 (1)	8.880 (2)	8.896 (1)	8.888 (1)	8.864 (1)	8.888 (2)	8.888 (1)	8.904 (2)
c_o (Å)	5.252 (2)	5.258 (2)	5.256 (1)	5.254 (1)	5.258 (1)	5.258 (1)	5.254 (1)	5.253 (1)
β (°)	106.32 (2)	106.24 (1)	106.15 (1)	106.20 (1)	106.39 (1)	106.16 (1)	106.21 (1)	106.15 (1)
V_{Cell} (Å^3)	432.96	435.33	436.67	435.68	433.56	436.29	435.59	437.10
N. Obs. Rifl.	565	640	613	601	600	573	576	406
R. Fin.	0.028	0.030	0.026	0.025	0.021	0.024	0.022	0.030
T-O1	1.610 (1)	1.610 (1)	1.607 (1)	1.608 (1)	1.613 (1)	1.610 (2)	1.605 (1)	1.605 (2)
T-O2	1.591 (2)	1.590 (2)	1.588 (2)	1.588 (1)	1.593 (2)	1.590 (2)	1.586 (2)	1.584 (3)
T-O3, A1	1.661 (2)	1.665 (2)	1.665 (2)	1.663 (1)	1.662 (1)	1.665 (2)	1.662 (1)	1.663 (3)
T-O3, A2	1.676 (3)	1.681 (3)	1.681 (2)	1.681 (1)	1.677 (1)	1.682 (2)	1.680 (2)	1.680 (5)
Mean	1.635 (4)	1.637 (4)	1.635 (4)	1.635 (2)	1.636 (2)	1.637 (4)	1.634 (3)	1.633 (7)
T-O brg.	1.669	1.673	1.673	1.672	1.669	1.674	1.671	1.671
T-O non brg.	1.601	1.600	1.598	1.598	1.603	1.600	1.596	1.595
V(T)	2.224 (2)	2.232 (3)	2.227 (2)	2.224 (1)	2.231 (2)	2.232 (1)	2.220 (1)	2.217 (4)
σ_θ^2tet.	25.19	25.06	26.06	25.30	24.37	25.47	25.48	25.32
M1-O2	2.020 (2)	2.031 (2)	2.039 (2)	2.035 (1)	2.019 (1)	2.036 (2)	2.036 (2)	2.050 (3)
M1-O1, A1	2.035 (2)	2.047 (2)	2.050 (1)	2.047 (1)	2.040 (1)	2.049 (1)	2.047 (1)	2.053 (3)
M1-O1, A2	2.109 (2)	2.114 (2)	2.118 (2)	2.114 (1)	2.112 (1)	2.118 (2)	2.118 (1)	2.122 (3)
Mean	2.054 (4)	2.064 (5)	2.069 (4)	2.066 (2)	2.057 (3)	2.068 (4)	2.067 (4)	2.075 (7)
V(M1)	11.458 (6)	11.624 (7)	11.714 (5)	11.654 (3)	11.502 (4)	11.692 (5)	11.673 (5)	11.820 (9)
σ_θ^2oct.	20.35	19.36	18.39	18.75	20.19	18.86	18.75	17.50
M2-O1	2.358 (2)	2.352 (2)	2.357 (2)	2.357 (1)	2.350 (1)	2.353 (2)	2.357 (1)	2.354 (3)
M2-O2	2.332 (2)	2.329 (2)	2.328 (2)	2.331 (1)	2.321 (1)	2.328 (2)	2.330 (1)	2.319 (3)
M2-O3, C1	2.540 (2)	2.553 (2)	2.558 (2)	2.556 (1)	2.550 (1)	2.559 (2)	2.554 (1)	2.568 (3)
M2-O3, C2	2.723 (2)	2.724 (2)	2.726 (2)	2.726 (1)	2.727 (1)	2.723 (2)	2.729 (2)	2.729 (4)
Mean	2.488 (5)	2.489 (5)	2.492 (5)	2.493 (3)	2.487 (3)	2.490 (5)	2.492 (4)	2.493 (9)
V(M2)	25.421 (10)	25.473 (10)	25.562 (10)	25.569 (5)	25.373 (6)	25.506 (10)	25.561 (8)	25.560 (16)
Δ M2	0.313	0.313	0.310	0.311	0.321	0.310	0.315	0.315

1996 and references therein). Note that Sr content (ppm) of the Nemby cpx (mean 619; range 38–2692) is on average distinctly higher than that for both the cpx from Mt. Leura (mean 163; range 80–352) and that from NE-Brazil (mean 110; range 12–287) (*Rivalenti* et al., 1996).

The present paper aims to investigate the crystal chemistry of the clinopyroxenes from Nemby spinel-peridotite xenoliths and its petrological implications.

Experimental procedures

X-ray diffraction data of inclusion-free crystals were recorded by means of an automated KUMA-KM4 (K-geometry) diffractometer, using MoKα radiation, monochromatized by a flat graphite crystal. Data collections were made up to 30 degrees θ with continuous ω-scan mode. Peak-base width is of 2° 2θ and counting times are from 20 to 40 s, as a function of peak σ. Background was measured for half peak-time. The correction for absorption was made according to *North* et al. (1968). 24 reflections (about 90° of 2θ) were accurately centred and used for cell parameter determination. Best results of structure refinement, performed with a SHELX-93 program (*Sheldrick*, 1993), were obtained using scattering curves (*International Tables for X-ray Crystallography*, 1974; *Tokonami*, 1965) for fully ionised Mg vs. Fe^{2+} and Ca vs. Na in M1 and M2 sites, respectively. $Si^{2.5+}$ and $O^{1.5-}$ scattering curves were used for T-site and the three oxygens, respectively. Final Fourier differences syntheses showed no significant residual density.

The same crystals used for X-ray single crystal data collection were utilised for microanalyses. These were performed by means of a CAMECA-CAMEBAX electron microprobe operating at 15 kV and 15 nA. A PAP-CAMECA program was used to convert X-ray counts into weight percent of the corresponding oxides. Results are accurate to 2–3% for major elements and better than 10% for minor elements.

Site occupancies were calculated using the equations of *Dal Negro* et al. (1982). The estimation of Fe^{3+} is based both on charge-balance and geometrical parameters, i.e. mean M1-O bond distance and M1 electrons, derived from the crystal structure refinements.

The results of the structure refinement, chemical composition and cation site partitioning are given in Tables 2 and 3, respectively.

Crystal chemistry

The Nemby cpx are characterised by high Ca (0.776–0.904 atoms per formula unit, a.f.u.) and Mg (0.834–0.942 a.f.u.) contents, and plot in the aluminous field of *Burnham* et al. (1967).

T-Site

In general, the T site volume (V(T)) decreases with decreasing Al^{IV} (Fig. 2A) and this is correlated with the increase of mg# cpx and the general decrease of cpx modal content (i.e. increase of basalt component extraction). Comparison between V(T) and cell volume (V(Cell)) shows (Fig. 2B) that the Nemby cpx have different

Table 3. *Chemical analyses and elemental cation partition among T, M1 and M2 sites for LK and HK cpx*

Sample	LK3192	LK3221	LK3227	LK3269	HK3311	HK3307	HK3211	HK3284
SiO_2	52.5	52.9	54.3	53.9	52.8	52.6	54.2	54.3
TiO_2	0.26	0.22	0.07	0.05	0.19	0.22	0.06	0.18
Al_2O_3	5.2	3.9	2.5	2.8	6.2	4.0	2.8	1.1
Cr_2O_3	0.9	0.8	0.8	0.6	0.8	0.8	0.8	0.4
FeO	2.7	2.7	2.2	2.1	2.9	2.5	2.4	2.9
MnO	0.01	0.06	0.03	0.00	0.13	0.07	0.02	0.07
MgO	15.1	16.4	17.1	16.7	15.5	15.6	16.6	17.5
CaO	21.2	21.5	22.6	22.1	20.1	22.9	21.4	23.4
Na_2O	1.7	1.2	0.9	1.2	1.5	1.0	1.5	0.6
Sum	99.57	99.68	100.50	99.45	100.12	99.69	99.78	100.45
T-Site								
Si	1.905	1.924	1.952	1.956	1.903	1.914	1.962	1.957
Al	0.095	0.076	0.048	0.044	0.097	0.086	0.038	0.043
Sum	2.000	2.000	2.000	2.000	2.000	2.000	2.000	2.000
Ml-Site								
Mg	0.776	0.833	0.871	0.857	0.743	0.827	0.844	0.901
Fe^{2+}	0.016	0.022	0.019	0.019	0.061	0.024	0.016	0.019
Al	0.128	0.091	0.059	0.077	0.167	0.086	0.083	0.006
Fe^{3+}	0.047	0.026	0.027	0.027	0.001	0.033	0.032	0.057
Cr	0.026	0.022	0.022	0.018	0.023	0.024	0.023	0.012
Ti	0.007	0.006	0.002	0.001	0.005	0.006	0.002	0.005
Sum	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000
M2-Site								
Ca	0.822	0.837	0.871	0.857	0.776	0.892	0.828	0.903
Na	0.120	0.075	0.063	0.081	0.104	0.069	0.102	0.042
Mg	0.040	0.053	0.045	0.044	0.091	0.018	0.046	0.040
Fe^{2+}	0.018	0.033	0.020	0.018	0.025	0.019	0.023	0.013
Mn	0.000	0.002	0.001	0.000	0.004	0.002	0.001	0.002
Sum	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000

V(Cell) for similar V(T), independently if it belongs to the LK and HK suite. Note that the Mt. Leura cpx show V(Cell) distinctly lower than those of most cpx from Nemby and NE-Brazil.

M2-Site

Ca (0.776–0.904 a.f.u.), and Na (0.120–0.042 a.f.u.), Mg (0.018 – 0.091 a.f.u.) and Fe^{2+} (0.011–0.033 a.f.u.; Table 3) essentially fill M2 site. The M2 site volume (V(M2)) of Nemby cpx parallels Ca increase, while those for Mt. Leura and NE-Brazil cpx show small variations or are virtually constant (Fig. 3A). This is due to the Al^{IV} and R^{3+} relationship where the charge balance depends on $Al^{IV} + Na$ vs. R^{3+}, Al^{IV} vs. R^{3+} and Na vs. R^{3+}, in Nemby, Mt. Leura and NE-Brazil cpx, respectively.

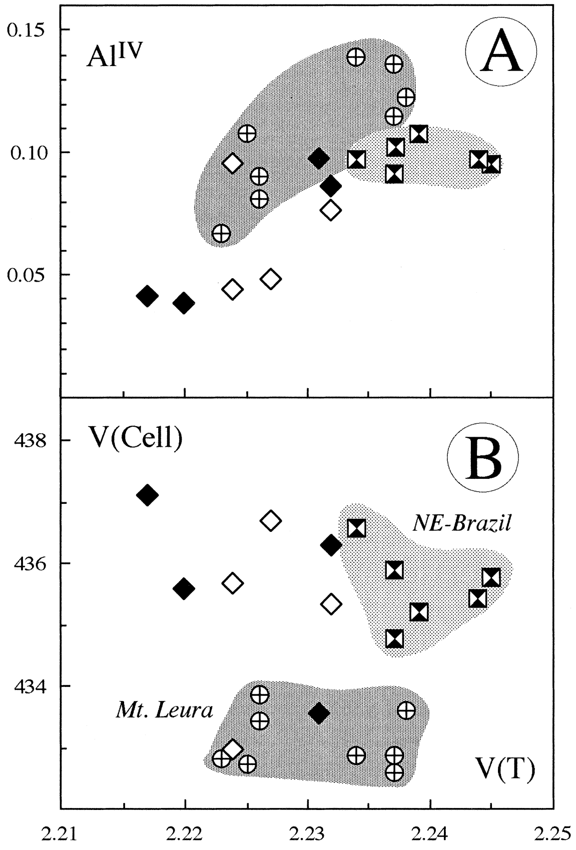

Fig. 2. V(T) vs. Al^{IV} content (**A**) and vs. V(cell) (**B**). Symbols as in Fig. 1

V(Cell) and V(M2) variations (Fig. 3B) indicate that two Nemby (LK3192 and HK3311) and Mt. Leura cpx have the lowest V(Cell) associated with the lowest V(M2), relative to the other Nemby and NE-Brazil cpx.

M1-Site

Mg (0.743–0.901 a.f.u.) and R^{3+} (Al^{VI}, Fe^{3+}, Cr and Ti^{4+}; 0.080–0.208 a.f.u.) cations fill M1 polyhedron where Fe^{2+} is present only in small amount (0.016–0.061 a.f.u.; Table 3). R^{3+} decrease, particularly Al^{VI}, controls the linear increase of the M1 volume (V(M1)) (Fig. 4A) to which conform both HK and LK Nemby cpx suites. It should be noted that Nemby and NE-Brazil cpx with the largest V(Cell) are also characterised by the largest V(M1) with respect to the LK3192 and HK3311 Nemby and Mt. Leura cpx (Fig. 4B).

Discussion and conclusions

As previously stated, crystal chemistry indicates that both LK and HK cpx from Nemby are characterised, for similar V(T), by different V(M1), V(M2) and V(Cell). This may indicate variable equilibration pressures in the peridotite spinel-facies (*Dal Negro* et al., 1984; *Princivalle* et al., 1989).

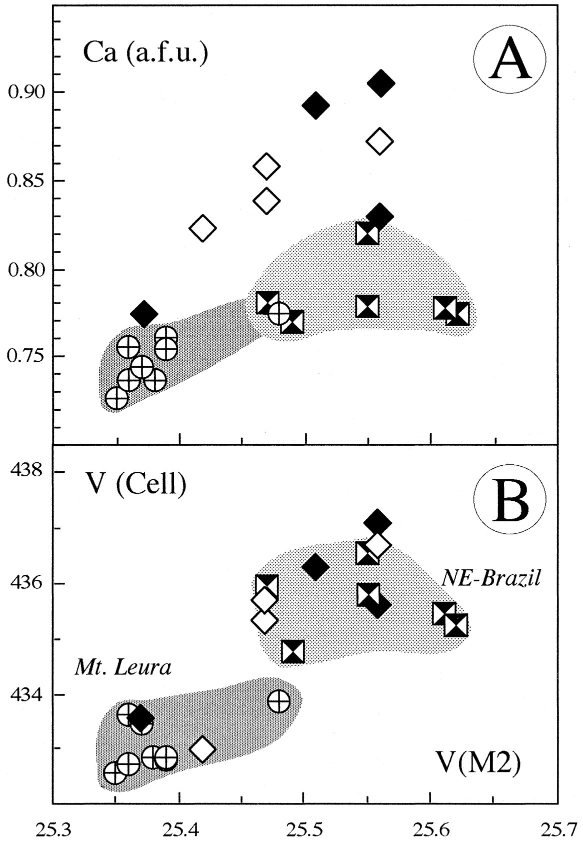

Fig. 3. V(M2) vs. Ca content (**A**) and vs. V(cell) (**B**). Symbols as in Fig. 1

According to *Mercier* (1980), the Nemby cpx equilibration pressure ranges from 12 to 20 kb. Clinopyroxenes from NE-Brazil and Mt. Leura mantle xenoliths yielded equilibration pressure ranging from 17 to 24, and from 15 to 20 kb, respectively (*Princivalle* et al., 1994, 1995). This supports that the LK and HK Nemby xenoliths were sampled in the same pressure range as the Mt. Leura and NE-Brazil xenoliths.

As suggested by *Dal Negro* et al. (1984, 1989), *Cundari* et al. (1986) and *Princivalle* et al. (1989) for cpx belonging to spinel peridotites depleted by basalt extraction, as well as for cpx crystallised from basic magmas at various pressure conditions (*Nimis* and *Ulmer*, 1998 and references therein), the decrease of both V(cell) and V(M1) is due to pressure increase. The variations of V(Cell) and V(M1) of cpx from mantle peridotite xenoliths, equilibrated between plagioclase-spinel to spinel-garnet facies (i.e. 9–12 and 18–22 kb, respectively; *Wilson*, 1996), are illustrated in Fig. 4B. The lowest V(Cell) and V(M1) refer to cpx of spinel-garnet xenoliths, while the highest volumes are relative to cpx from plagioclase-spinel peridotites. The investigated Nemby cpx have V(Cell) and V(M1) which are intermediate between those of plagioclase- and garnet-bearing mantle peridotites.

In conclusion, V(Cell) and V(M1) relationships are more sensitive than the only chemical composition, to estimate the equilibration pressure of cpx (*Princivalle* et al., 1994).

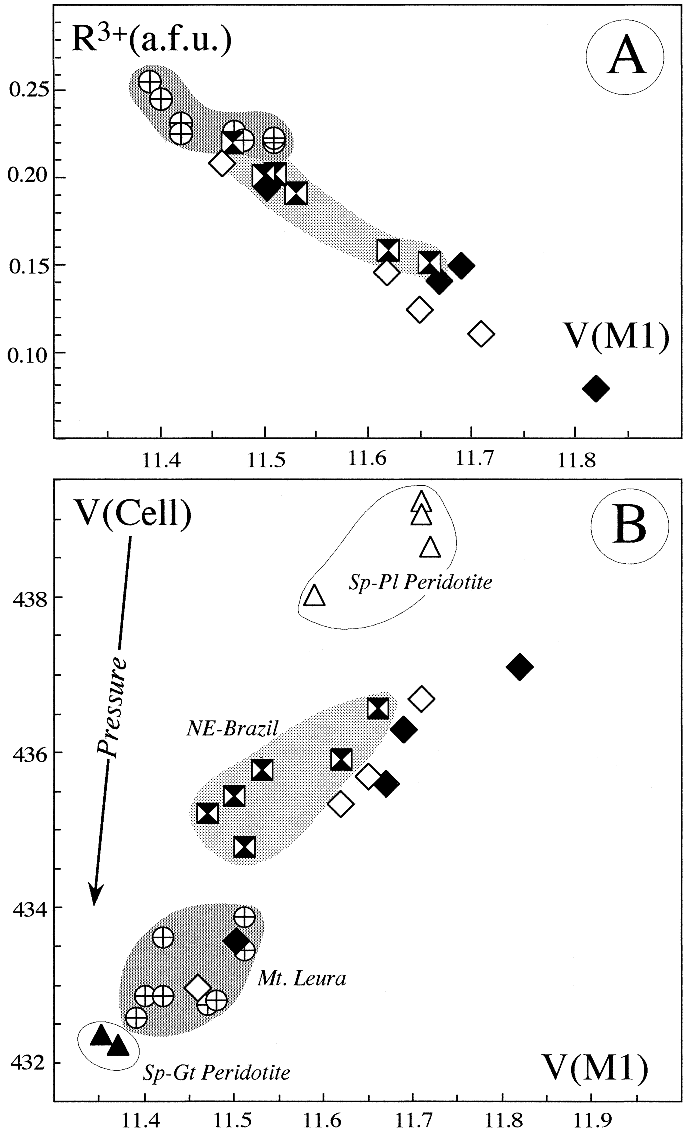

Fig. 4. V(M1) vs. R^{3+} content (**A**) and vs. V(Cell) (**B**). Symbols as in Fig. 1. Solid triangles: cpx from spinel-garnet (Sp-Gt) peridotite (Tasmania; *Nimis*, 1995), triangles: cpx from spinel-plagioclase (Sp-Pl) peridotite (Zabargard; *Nimis*, 1995)

The Nemby xenoliths represent mantle peridotites sampled in a large range of pressure (ca. 12–18 kb). It should be noted that the influence of the "metasomatic processes", even if very pronounced as for the HK Nemby xenoliths, on the cpx polyhedral variations is not appreciable.

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