*American Mineralogist, Volume 95, pages 196–199, 2010*

**Letter**

# Corundum + orthopyroxene ± spinel intergrowths in an ultrahigh-temperature Al-Mg granulite from the Southern Marginal Zone, Limpopo Belt, South Africa

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## Abstract

We report the occurrence of unusual lamellar (random and regular subparallel) intergrowths of corundum and symplectic intergrowth of spinel with orthopyroxene in an ultrahigh-temperature Al-Mg granulite from the Southern Marginal Zone of the Limpopo Belt, South Africa. Separate compositions are suggested for the homogenous phase that might have existed prior to the formation of the two types of lamellar intergrowths of corundum with orthopyroxene. In the case of random lamellar corundum + orthopyroxene intergrowth, the estimated precursor composition is similar to Fe-Mg garnet, while, although speculative, an ultrahigh-Al orthopyroxene precursor is suggested to account for the exsolution nature of the unique regular subparallel corundum lamellae in orthopyroxene. Considering that the stability field of Fe-Mg-rich garnet relative to orthopyroxene + corundum extends to lower pressures, the estimated garnet composition ($X_{Mg}$ = ~0.65) is most likely stable at low pressures of ~5 kbar. Such low pressures are further supported by the Fe-Mg-rich garnet compositional nature of the pre-spinel-intergrowth orthopyroxene. Given the rare preservation of the mineral assemblages typical of prograde metamorphism in granulite facies rocks, our discovery of corundum lamellar intergrowth with orthopyroxene from an ultrahigh-temperature Al-Mg granulite is unique.

**Keywords:** Corundum + orthopyroxene intergrowth, ultrahigh-temperature Al-Mg granulite, Fe-Mg-rich garnet, Limpopo Belt

## Introduction

Mineral compositions that are stable at high temperature or pressure sometimes become unstable as a result of cooling to low temperatures or decompression to low pressures. The resulting textures (symplectic and/or lamellar intergrowth of two minerals) reflect equilibrium and/or non-equilibrium thermodynamic information that can potentially be used to infer earlier pressure and temperature conditions. This is especially useful in the case of granulite facies rocks where mineralogical and mineral chemical evidence for prograde metamorphism is rarely preserved due to high diffusion and high reaction rates that erase evidence of the prograde history (Harley 2004; Kelsey 2008). In this study, we report on the occurrence of lamellar intergrowth of corundum and symplectic intergrowth of spinel with orthopyroxene in an Al-Mg granulite from the Southern Marginal Zone of the Limpopo Belt. Belyanin et al. (in review) recently reported ultrahigh-temperature conditions for this same granulite. Our report is significant considering that granulites that preserve the near-peak mineral assemblage orthopyroxene + corundum are rare (Kelly and Harley 2004 and references therein), and that corundum lamellar intergrowth with orthopyroxene has not been previously reported from granulite-facies terrains.

## Geologic setting

The Limpopo Belt of South Africa (Fig. 1), situated between the Archean Kaapvaal Craton to the south and the Zimbabwe Craton to the north, is a structurally complex high-grade metamorphic province that has an exposed strike length of about 700 km and a width of about 200 km. The three domains, the Northern Marginal Zone, Central Zone, and Southern Marginal Zone, are separated from each other and also from the surrounding cratons by prominent shear zones. The Northern and Southern Marginal Zones, which are composed of reworked rocks of the respective cratons, are thrust onto the adjacent cratons along >10 km wide steep inward-dipping shear zones at ~2.65 Ga, the North Limpopo Thrust Zone and the Hout River Shear Zone, respectively (Kramers et al. 2006 and references therein). Available geochronologic, structural, and petrologic data have been interpreted by various researchers to show that whereas all three sub-zones have been

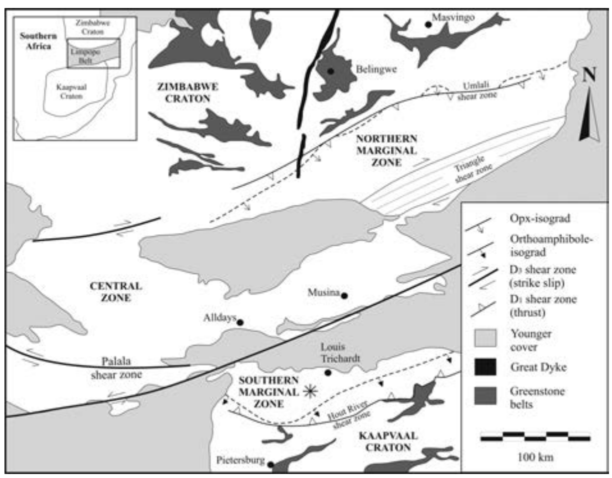


**Figure 1.** Generalized geological map of the Limpopo Belt showing the three sub-zones and the location of the granulite studied here (shown by star), about 15 km NW of the Bendelierkop quarry.

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0003-004X/10/0001–196$05.00/DOI: 10.2138/am.2010.3383 

affected by a major Neoarchean high-grade tectono-metamorphic event within the same time interval (~2.6–2.5 Ga), the Central Zone alone was again reworked in the Paleoproterozoic (~2.0 Ga) (Van Reenen et al. 2008 and references therein).

The Southern Marginal Zone exposes volumetrically dominant tonalitic to trondhjemitic gray gneiss (the Goudplaats/Hout River Gneiss) intercalated with granulite facies supracrustal gneiss of mafic, ultramafic, and metapelitic compositions (the Bandelierkop Complex), closely associated with metamorphosed banded iron formations, and late granitoid intrusions (e.g., Du Toit et al. 1983). The timing of granulite facies metamorphism in the Southern Marginal Zone is constrained to 2691 ± 7 Ma by a U-Pb date on monazite from a metapelitic granulite exposed at the Bandelierkop quarry (Kreissig et al. 2001). Although the metamorphic evolution of the Southern Marginal Zone was a matter of investigation for the last two decades, the ultrahigh-temperature event was not reported until recently. Tsunogae et al. (2004) reported ultrahigh-temperature conditions of ~950 °C from the Southern Marginal Zone using early compositions of antiperthite in quartzo-feldspathic gneiss and high-Al (~9 wt%) orthopyroxene in pelitic granulite. Belyanin et al. (in review) estimated higher temperatures ($T \geq 1050$ °C at ~10 kbar) and presented clear textural evidence and mineral assemblages [orthopyroxene + sillimanite ± quartz symplectites, Al-rich orthopyroxene (~11 wt%), antiperthites, spinel + quartz, corundum + quartz, and orthopyroxene + ilmenite] typical of ultrahigh-temperature conditions from an Al-Mg granulite. The unique textures presented here are from the same ultrahigh-temperature granulite exposed 15 km northwest of the Bendelierkop quarry (Fig. 1) (Van Reenen 1983), forming part of the Bandelierkop Complex in the Southern Marginal Zone.

## Petrography and mineral compositions

The granulite studied has a gneissic fabric characterized by distinct Al-rich layers alternating with Si-rich layers and is medium grained and porphyroblastic. The rock has low Ca and Si contents, and moderate Al content ($SiO_2$ ~56.85 wt%, $TiO_2$ ~0.87 wt%, $Al_2O_3$ ~20.04 wt%, $Fe_2O_3$ ~1.25 wt%, FeO ~7.14 wt%, MgO ~6.86 wt%, CaO ~0.78 wt%, $Na_2O$ ~1.26 wt%, $K_2O$ ~2.67 wt%; Van Reenen 1983). Electron microprobe analyses of minerals in representative thin sections from the granulite were carried out using a CAMECA SX100, equipped with an EDS detector, housed at the central analytical facility (SPECTRAU) of the University of Johannesburg. Analytical conditions of 15 kV accelerating voltage, 20 nA beam current, counting times of 10–20 s, and 1–5 μm beam spot size were used. Representative chemical compositions of corundum + orthopyroxene ± spinel intergrowths are given in Table 1[1].

We focus on the Al-rich layer where the unique and spectacular textures reported here are observed. This layer is dominantly composed of sillimanite, cordierite, spinel, orthopyroxene, biotite, and quartz, with rare garnet, corundum, and staurolite. Accessory phases include ilmenite, pyrite, pentlandite, and rutile. The texture that attracts our attention is the occurrence of orthopyroxene + sillimanite ± quartz symplectites adjacent to or surrounding cordierite and/or sillimanite grains. Rare corundum occurs as elongate sub-idiomorphic grains in association with ilmenite, spinel, cordierite, and sillimanite (Figs. 2a and 2b), as large grains in association with pyrite and/or spinel (Fig. 2c), as small altered xenomorphic inclusions in spinel (Figs. 2a and 2b), and as lamellar intergrowths with orthopyroxene (Figs. 2b, 2c, 2d, 2e, and 2f). In terms of composition, the small altered xenomorphic corundum grains (Figs. 2a and 2b) have higher Cr and Fe contents than the sub-idiomorphic or large corundum grains (Figs. 2a, 2b, 2c, and 2d) [Cr/(Cr + Al) = 0.012 and 0.004, respectively; FeO = 1.52 and ~0.50 wt%, respectively; Table 1[1]]. In addition to the large grains (Figs. 2a, 2b, 2c, 2d, and 2f), spinel also occurs as a symplectite with orthopyroxene (Figs. 2b, 2c, and 2f) and as inclusions in large corundum grains (Fig. 2c). Spinel inclusions in corundum (Fig. 2c) are characterized by their higher $Cr_2O_3$ (~10 wt%) contents and lower $X_{Mg}$ [Mg/(Mg+$Fe^{2+}$) = ~0.29] relative to the large spinel grains (Figs. 2a, 2b, 2c, 2d, and 2f) ($Cr_2O_3$ = ~0.98 wt%, $X_{Mg}$ = 0.38) (Table 1). Staurolite inclusions in corundum are characterized by their higher $TiO_2$ (~0.98 wt%) and $X_{Mg}$ (~0.34) relative to staurolite inclusions in spinel (~0.25 wt% and ~0.3) and orthopyroxene (~0.06 wt% and ~0.32).

Two types of lamellar intergrowths of corundum ± spinel with orthopyroxene grains are observed and are characterized by either: (1) a dominant region of elongate randomly oriented corundum lamellae and a relatively small region of orthopyroxene + spinel symplectic intergrowth (Figs. 2b and 2f), or (2) a dominant region of orthopyroxene + spinel symplectic intergrowth, an intermediate region of orthopyroxene + spinel symplectic intergrowth and corundum lamellae, and a relatively small region of elongate regular subparallel corundum lamellae in orthopyroxene (Fig. 2c). Rarely, orthopyroxene grains show only randomly oriented corundum lamellae (Fig. 2d). Based on their arrangement in orthopyroxene, the randomly oriented corundum lamellae (Figs. 2b, 2d, and 2f) appear to be of the same paragenesis with orthopyroxene (intergrowth), whereas the regular subparallel corundum lamellae appear to be a product of exsolution (Fig. 2c). Although the different orthopyroxene grains have similar $X_{Mg}$ (~0.62–0.64), orthopyroxene coexisting with random corundum lamellae has higher Al content (~7.3 wt%) than those coexisting with regular subparallel corundum lamellae (~5.9 wt%) or spinel (~6.49–6.72 wt%) (Table 1).

The reintegrated composition of both the orthopyroxene-corundum (random lamellar) intergrowth (measured using a defocused beam with 40 μm spot size) (Figs. 2b and 2f) and the orthopyroxene-spinel intergrowth (measured using a defocused beam with 15 to 20 μm spot size) (Fig. 2c) is similar to Fe-Mg-rich garnet, with the latter having a higher $X_{Mg}$ and lower $Al_2O_3$ (~0.60 and 27 wt% and ~0.62–0.65 and 18–21 wt%, respectively; Table 1). The Fe-Mg-rich garnet composition of the pre-random-lamellar-corundum intergrowth phase is further supported by the presence of small Fe-Mg-rich garnet inclusions in orthopyroxene within sillimanite (Fig. 2d). Because of their small size, we used energy-dispersive X-ray maps to delineate and identify the phases (Fig. 2e). Analytical conditions of 15 kV acceleration voltage, 40

[1] Deposit item AM-10-003, Table 1. Deposit items are available two ways: For a paper copy contact the Business Office of the Mineralogical Society of America (see inside front cover of recent issue) for price information. For an electronic copy visit the MSA web site at http://www.minsocam.org, go to the *American Mineralogist* Contents, find the table of contents for the specific volume/issue wanted, and then click on the deposit link there.

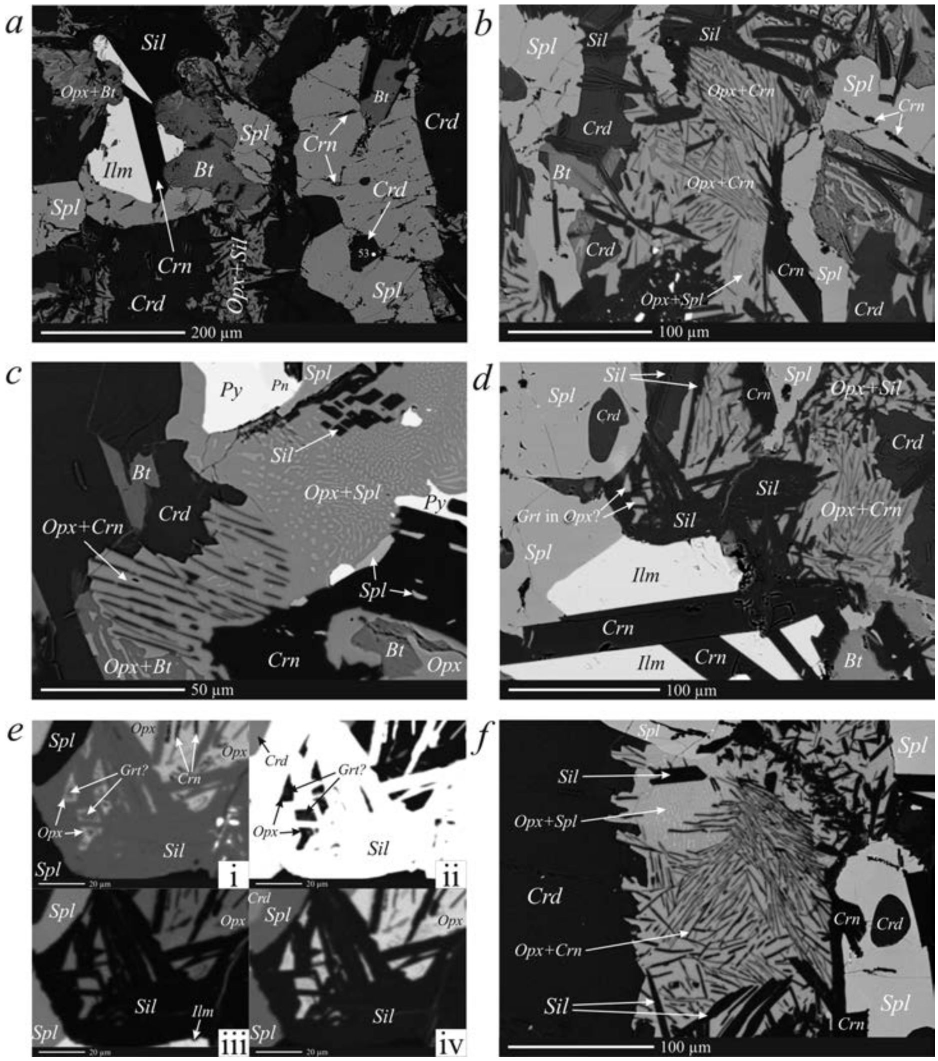


**FIGURE 2.** Representative back-scattered electron images illustrating (**a**) elongate sub-idiomorphic corundum in association with spinel, ilmenite and sillimanite, near a large spinel grain with small altered xenomorphic corundum; (**b**) sub-idiomorphic corundum in association with spinel, small altered xenomorphic corundum within spinel, and orthopyroxene showing randomly oriented corundum lamellae and symplectic intergrowth with spinel; (**c**) orthopyroxene showing regular subparallel corundum lamellae and symplectic intergrowths with spinel, near a large corundum grain; (**d**) elongate corundum near orthopyroxene showing randomly oriented corundum lamellae; (**e**) EDS mapping of the tiny garnet inclusion in orthopyroxene within sillimanite, seen slightly left of the center of the image in **d**; and (**f**) orthopyroxene showing randomly oriented corundum lamellae and symplectic intergrowth with spinel. The elements mapped in each image in **e** are: (i) Si; (ii) Al; (iii) Fe; and (iv) Mg. Numbers correspond to analytical data given in Table 1[1].

nA beam current, counting times of 100 ms, and resolution of 512 × 384 were used for EDS mapping. Although the analyses of these small garnet inclusions are slightly non-stoichiometric, their composition ($X_{Mg}$ = ~0.63), with $Al_2O_3$ contents of ~21 to 27.5 wt%, is similar to that obtained from the reintegrated (random lamellar) corundum-orthopyroxene intergrowths and orthopyroxene-spinel intergrowths (Table 1[1]). Because of spurious Al contents in the case of orthopyroxene with a small region of regular subparallel corundum lamellae (Fig. 2c), we resorted to reintegrated composition using modal proportions (considering corundum as pure – 100% $Al_2O_3$) estimated with image-processing software. Details of reintegration are given in Table 1[1]. The pre-exsolution composition obtained is similar to orthopyroxene with an anomalously high alumina content (~21 wt% $Al_2O_3$; Table 1[1]).

## DISCUSSION

Although the corundum + orthopyroxene assemblage occurs as large crystals either in contact or separated from each other by sapphirine ± cordierite intergrowths in high-temperature Mg-Al granulites, no report exists on the intergrowth of these two minerals in the literature. Rare lamellar intergrowths of corundum with other minerals besides orthopyroxene are reported in the literature. For example, corundum laths in spinel and lamellar intergrowth of corundum and ilmenite have been reported from some granulite facies rocks (e.g., Moore 1971; Guiraud et al. 1996; Sengupta et al. 1999). In these cases, corundum lamellae are interpreted as being related to a decrease in the *P*-*T* conditions. The most similar example to that reported in this study is the elongated, oriented laths of corundum seen in clinopyroxene in corundum-bearing eclogite xenoliths from kimberlites (e.g., Viljoen et al. 2005). In spite of the occurrence of orthopyroxene in minor amounts (>1 vol%) in some eclogite xenoliths, no lamellar intergrowth of corundum + orthopyroxene has yet been reported.

In the granulite studied, the estimated composition of the homogenous phase that existed prior to random lamellar intergrowth with corundum is similar to Fe-Mg-rich garnet. This could be interpreted by the garnet breakdown reaction to form orthopyroxene and corundum (garnet = orthopyroxene + corundum). In the case of pure-Mg end-member garnet, this reaction lies at high pressures ≥15 kbar in the currently available *P-T* grid for the silica-undersaturated portions of the FMAS system (Gasparik and Newton 1984; Kelly and Harley 2004). However, for Fe-Mg-rich garnet, like in the granulite studied here, the stability of garnet Fe,Mg solid solutions relative to orthopyroxene + corundum extends to lower pressures because Fe partitions into garnet relative to orthopyroxene. Comparison with the garnet $X_{Mg}$ isopleths constructed by Kelly and Harley (2004) suggest that the composition of garnet ($X_{Mg}$ = ~0.65) in this granulite is stable at low pressures of ca. 5 kbar at 900 °C. Such low pressures (~5 kbar) are further supported by the Fe-Mg-rich garnet compositional nature of the pre-spinel-intergrowth orthopyroxene in the granulite studied here, as many well-documented occurrences of orthopyroxene + spinel intergrowths have developed by decompressive decomposition of garnet to orthopyroxene + spinel + cordierite at pressures of ~5 kbar (e.g., Droop and Bucher-Nurminen 1984).

On the other hand, the estimated composition of the homogeneous phase that existed prior to the exsolution of regular subparallel corundum lamellae is similar to orthopyroxene with an anomalously high alumina content (~21 wt%). Even though the maximum $Al_2O_3$ content reported so far in natural orthopyroxenes from aluminous granulites is ~13 wt% (Moraes et al. 2002; Sajeev and Osanai 2004) with Hollis and Harley's (2003) experimental study synthesizing an orthopyroxene with 15 wt% $Al_2O_3$, we note other reports on the rare Al-rich diopside ("fassaite") with extremely high (ultrahigh) $Al_2O_3$ (~16–21 wt%) in corundum-garnet-bearing xenoliths (e.g., Sutherland et al. 2003), and the rare Al-rich clinopyroxene, rich in Tschermak components and containing ultrahigh $Al_2O_3$ contents (~18–23 wt%), in Mg-Al sapphirine-bearing granulite xenoliths (e.g., Ulianov and Kalt 2006). Orthopyroxenes with ultrahigh content of alumina (~21 wt%), even at the extreme temperatures, have not been reported in aluminous granulites, nevertheless, they may be possible given the texture reported here.

The rare assemblage of orthopyroxene and corundum, predicted by chemographic and topological requirements and experimental studies (e.g., Waters 1986; Hensen 1987), and inferred from studies on reaction textures and inclusion assemblages (Morse and Talley 1971; Grew 1983; Ouzegane et al. 2003), was recently shown to occur as coexisting grains having reached textural equilibrium in a Mg-Al granulite from east Antarctica (Kelly and Harley 2004). Thus our discovery of corundum lamellar intergrowth with orthopyroxene from an ultrahigh-temperature Al-Mg granulite further helps improve understanding of Al-Mg granulites, a rare group of rocks in nature, which often preserve mineral assemblages that are diagnostic of extreme thermal regimes in the Earth's crust (Harley 2004; Kelsey 2008).

## Acknowledgments

A post-graduate grant of the Faculty of Science [University of Johannesburg (UJ)] provided financial assistance to the first author. Fieldwork and analytical expenses were supported by a Faculty of Science (UJ)/SASOL grant to H.M.R. D.D.v.R. acknowledges an NRF grant (GUN 2053192). Thought-provoking discussions with Toshiaki Tsunogae and Kaushik Das are much appreciated. The journal reviewers, M. Santosh and Martin Rigby, provided positive and encouraging comments. This is a contribution to Paleoproterozoic Mineralization (PPM) Research Group and Gondwana Institute for Geology and Environment.

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Manuscript received August 25, 2009
Manuscript accepted September 8, 2009
Manuscript handled by Bryan Chakoumakos