POLYGONAL SERPENTINE IN SEGREGATION-TEXTURED KIMBERLITE

ROGER H. MITCHELL

Department of Geology, Lakehead University, Thunder Bay, Ontario P7B 5E1

ANDREW PUTNIS

Department of Earth Sciences, University of Cambridge, Downing Street, Cambridge, England CB2 3EQ

ABSTRACT

Serpentine aggregates occurring in calcite-"serpophite" segregations in kimberlites are shown by electron optical methods to consist principally of polygonal serpentine accompanied by minor chrysotile and lizardite. The polygonal serpentine forms complex fibers consisting of sectors of flat layers having clinochrysotile-like stacking overlying a cylindrical chrysotile core. The serpentines exhibit a wide range of morphology and are considered to be primary phases that crystallized over a wide range of temperatures (600-400°C) from the residual fraction of kimberlite magma.

Keywords: serpentine, "serpophite", polygonal serpentine, chrysotile, kimberlite.

SOMMAIRE

Les agrégats de serpentine dans les ségrégations de calcite + "serpophite" des kimberlites, examinés par techniques de microscopie électronique, contiennent surtout de la serpentine en plaquettes polygonales, avec un peu de chrysotile et de lizardite. La serpentine polygonale est faite de fibres complexes à secteurs de feuillets plats ayant un empilement semblable à celui du clinochrysotile, qui recouvrent un noyau de chrysotile cylindrique. Les serpentines présentent une grande variété de développements morphologiques. Elles seraient des phases primaires cristallisées à partir de la fraction résiduelle du magma kimberlitique, dans un intervalle de 600 à 400°C.

(Traduit par la Rédaction)

Mots-clés: serpentine, "serpentine", serpentine polygonale, chrysotile, kimberlite.

INTRODUCTION

Serpentine is one of the dominant groundmass minerals of kimberlite, ranging in modal abundance from 20 to 50% (Skinner & Clement 1979). Mitchell (1986) has noted that serpentine occurs in three parageneses in kimberlite: (1) as pseudomorphic replacements of earlier minerals such as olivine, (2) as irregular segregations of apparently primary serpentine in the groundmass, and (3) as nonpseudomorphic replacements of pre-existing pseudomorphic serpentine of paragenesis 1. Segregation serpentine, which is the subject of this paper, occurs as extremely fine-grained homogeneous pale green or brown amorphous-appearing masses that are typically isotropic or very weakly birefringent. Such serpentines are commonly termed *serpophite* (Lodochnikov 1933, Varlakov & Guryev 1985). This name is a useful descriptive term for uncharacterized serpentine but has no strict mineralogical meaning. Wicks & Zussman (1975) and Wicks & Whittaker (1977) have shown that *serpophite* is not amorphous but rather an extremely fine-grained aggregate of identifiable minerals.

Despite the ubiquity of serpentine in kimberlites, few studies have attempted to identify the polymorphs and polytypes present. The majority of the existing data, summarized by Mitchell (1986), refers to serpentine found in paragenesis 1 or to X-ray studies of whole-rock powders. Lizardite, antigorite and chrysotile have been reported to be present.

X-ray microdiffraction (Wicks & Zussman 1975) studies of serpophitic serpentine by Jago & Mitchell (1985) indicated the presence of lizardite-IT. Investigations by Kornilova et al. (1981), Podvysotskiy et al. (1987) and Podvysotskiy (1985) suggested that almost all groundmass serpophite is lizardite. Electron-microscope observations have shown that in some of these cases, lizardite has a "rounded edge", which Podvysotskiy (1985) interpreted as partial conversion to chrysotile. X-ray-diffraction studies (Podvysotskiy 1985) gave diffraction patterns that suggest the presence of a polygonal serpentine intermediate in structure between chrysotile and lizardite. Such serpentine was initially recognized from metamorphosed ultrabasic rocks by Krstanovič & Pavlovič (1964, 1967) and was described as Povlen-type serpentine.

Middleton & Whittaker (1976) proposed that Povlen-type serpentine be termed polygonal serpentine to reflect its structure, which consists of polygonally arranged flat layers of lizardite or chrysotile around a cylindrical core of chrysotile. This polygonal arrangement of serpentine polymorphs is an important and common form of serpentine in metamorphic rocks (Cressey 1979, Cressey & Zussman 1976, Jiang & Liu 1984, Mellini 1986) but has not been reported from an igneous paragenesis.

The segregations studied here consist entirely of

serpophitic serpentine or are a mixture of coarsely crystalline calcite and serpentine. Textural evidence suggests that calcite crystallized before the serpentine and, in many examples, became partially resorbed prior to serpentine deposition. Mitchell (1986) suggested that the segregations represent the last phase of kimberlite crystallization. The segregation seems to be related to surface-tension effects between the residual MgO-SiO₂-H₂O-rich fluid and the plastic, suprasolidus, silicate-oxide-bearing groundmass. The segregations are not the result of liquid immiscibility. The serpophitic serpentine comprising the segregations apparently is a primary assemblage of late-stage, low-temperature minerals.

EXPERIMENTAL METHODS

Samples of hypabyssal kimberlite from the Elwin Bay and Ham kimberlites, Somerset Island, Northwest Territories, and the Frank Smith kimberlite, South Africa, were utilized in the investigation. Kimberlites containing serpophitic segregations up to 2 mm in size were prepared as standard uncovered petrographic thin sections mounted using Crystal Bond[®].

Scanning electron microscope (SEM) studies were undertaken on gold-coated thin sections that had been etched for 2 minutes in 5% (vol.) HCl. Observations were made using a Philips 5015M SEM at



FIGS. 1-4. (1) Detached bundle of a subparallel growth-aggregate of smooth polygonal serpentine. (2) Irregular corallike aggregates of polygonal serpentine. (3) Polygonal serpentine consisting of three growth-zones. (4) Aggregate of polygonal serpentines illustrating their complex internal tube-in-tube morphology.

the University of Cambridge and a Hitachi 750 SEM at Lakehead University.

Transmission electron microscope (TEM) studies were done on ion-milled samples that had been cut from thin sections and mounted on copper grids. Samples were not etched. Low-resolution studies were made using a Philips 400T electron microscope, and high-resolution lattice images were obtained with a JEOL JEM 100CX instrument at the University of Cambridge. The TEM imaging was handicapped by beam-damage effects, causing the serpentine samples to become amorphous within a few seconds of exposure to the electron beam.

RESULTS

Scanning electron microscopy

The morphology of the serpophitic serpentines as revealed by SEM study is extraordinarily complex. The segregations consist principally of relatively large fibers of serpentine that are $0.5-1 \mu m$ in diameter and up to 30 μm long. Where calcite has been etched from the segregation it is clear that the fibers comprising the serpophite are stacked in subparallel aggregates (Fig. 1). Intergrown with the large fibers are curved ones that are less than 0.1 μm in diameter



FIGS. 5-8. (5) Decorated polygonal serpentines. (6) Decorated chrysotile serpentines.(7) Detail of plate-like decorations at the terminations of polygonal serpentines at the margin of an etched segregation. (8) A substrate of serpentine with globular morphology on the wall of an etched segregation.



FIG. 9. Low-resolution electron micrograph of polygonal serpentine. Note the hollow core of chrysotile visible in transverse and longitudinal sections.



FIG. 10. Selected-area electron-diffraction pattern of polygonal serpentine.

but are of similar length to the large fibers. These thin fibers are morphologically similar to chrysotile fibers.

Other segregations contain large (up to 1 μ m diameter) irregular fibers that occur as randomly intergrown masses or as coral-like sprays originating from the segregation wall (Fig. 2). Both varieties nucleate on substrates of subparallel aggregates of serpentine fibers.

The subparallel large fibers typically have smooth

exterior surfaces. Figures 3 and 4 show that each fiber is composed internally of 2 to 3 concentric tubes. Similar tube-in-tube structures have previously been reported in TEM studies of synthetic and natural serpentines (Bates 1961, Jiang & Liu 1984).

The irregular large fibers (Fig. 2) are typically decorated by small irregular plates (Fig. 5) or by globular and colloform structures (Figs. 5, 6). Intergrown thin curved chrysotile fibers are similarly, though not as extensively, decorated (Fig. 6).

The plate-like and globular structures also occur as mound-like aggregates upon calcite or platy pseudomorphic lizardite substrates (Figs. 7, 8). The globular structures display fractal properties with respect to the degree of magnification, suggesting that formation involved random nucleation.

Electron petrography

Low-resolution TEM of serpophitic serpentines (Fig. 9) illustrate the internal structure of the large fibers. Sections cut normal to the fiber axis exhibit a hollow core and a circular cross-section. Radial extinction-contours originate from this tubular core. The number of polygonal sectors was estimated to be approximately 30 per fiber, although accurate counts could not be made as a result of beam damage. Sections cut parallel to the fiber axis show the central tube as an area of differing degrees of diffraction contrast to the mantling material.

Interspersed with these tubular fibers are small isolated cylindral fibers of chrysotile together with rarer large laths of material lacking a central tube. These larger laths show greater diffraction-contrast than longitudinal sections of the tubular fibers and have been identified by electron diffraction as lizardite.

Electron diffraction

Electron-diffraction patterns of the fibers demonstrate the presence of dominant clinochrysotile and minor orthochrysotile. Figure 10 is an example of a pattern obtained with the electron beam normal to the fiber axis. The spacing between the rows (or layers) of reflections represents the a repeat-distance of 5.3 Å along the fiber axis. The equal intensities of each member of pairs of 00l and 0k0 reflections along the zero-layer line (h = 0) indicate a cylindrical lattice (Zussman et al. 1957) with Z arranged in all positions about the circumference of the fiber and perpendicular to the fiber axis. Y is contained within the 7-Å layer and is concentrically curved about the fiber axis. The hk0 reflections on the first layer-line (h = 1) are streaked along the layer line and are interpreted to indicate regular stacking of the layers along Z, coupled with disorder along Y (Whittaker & Zussman 1971). The c repeat, as determined from the 00l reflections, is 7.2 Å. Intensity nodes on the tails of diffuse streaks can be indexed as *hkl* reflections from polygonal material (Middleton & Whittaker 1976).

Reflections indexed as 20/ of clinochrysotile are located on a clinonet; those on an orthonet are 20/ of orthochrysotile. Streaking in these and higherorder even layers is related to disordered stacking along Y.

The electron-diffraction data clearly demonstrate that the fibers exhibiting radial polygonal sectors have chrysotile-like stacking.



FIG. 11. Lattice image of polygonal serpentine. Note the noncoherent antigoriteoffsets in area A, which grade into coherent chrysotile polygonal plates in area B.



FIG. 12. Lattice image of polygonal serpentine showing three growth layers. Note the strongly curved 7.2-Å layers in areas 1 and 3.

High-resolution microscopy

The identification of polygonal serpentine in the segregations is confirmed by high-resolution TEM images (Figs. 11, 12), in which the 7.2 Å interplanar spacing of the 1:1 layer silicate is resolved.

Figure 11 shows part of a cross-section of a fiber consisting of flat plates arranged according to the model of Middleton & Whittaker (1976) for polygonal serpentine. The sector boundaries are, for the most part, coherent with respect to the continuation of the 1:1 layers across the boundary, with angular changes of 15°. Area A (Fig. 11) contains plates with noncoherent boundaries. This region grades outward to coherent plates (Fig. 11, area B). The discontinuities in the 1:1 layer are similar to the "antigorite offsets" described and explained by Livi & Veblen (1987) as a reversal in the orientation of the octahedral and tetrahedral layers with respect to the adjacent plate. In this polygonal serpentine, the feature appears to be related to the growth of successive layers, but the cause of the reversal and of the subsequent return to coherency is unknown. This feature has not been recognized previously in polygonal serpentine.

Figure 12 shows a complex fiber consisting of three units. A central core (1) composed of strongly curved layers is overlain by a fiber (2) composed of polygonal sectors. An outer fiber (3) of complex structure mantles fiber 2. Fibers 2 and 3 exhibit different degrees of beam damage, suggesting that they have slightly different compositions or structures (or both).

Polygonal plates in fiber 3 are in general arranged parallel to plates in fiber 2, in a manner suggestive of crystallographic control of growth. A major growth-defect or discontinuity, however, is present in one area of fiber 3. Here, fiber-3 plates transform into strongly curved layers that terminate approximately normally to the exterior surface of fiber 2. The structure is not the result of superposition of two fibers, as the curved layers are continuous with the polygonal plates of the sectors. Similar curved features have been documented previously by Veblen & Buseck (1981).

CONCLUSIONS

The above observations demonstrate that serpophitic segregations in kimberlite consist principally of large polygonal serpentine fibers associated with lesser amounts of thin chrysotile tubes and plates of lizardite-1T. Polygonal serpentine seems to have grown upon pre-existing substrates of chrysotile. The complex morphology of the polygonal fibers (Fig. 12) suggests that serpentine crystallization was not continuous, and that up to three periods of crystallization were involved in the growth of some fibers. Polygonal serpentine and chrysotile are commonly overgrown by further platy and globular serpentines.

Mitchell (1986) suggested that calcite-serpentine segregations form over the temperature range 600-800°C. This study indicates that serpentine crystallized over this temperature interval and suggests that different polymorphs and morphologies may be deposited at particular temperatures.

Middleton & Whittaker (1976) recognized two varieties of polygonal serpentine: (1) orthochrysotile cylindrical cores with overlying lizardite polygonal sectors, and (2) clinochrysotile cylindrical cores with polygonal sectors consisting of flat layers having clinochrysotile-like stacking. The electron-diffraction data suggest that the polygonal serpentine in the segregations corresponds to the latter variety.

In summary, this study has demonstrated the importance of polygonal serpentine as a constituent of serpophite segregations in kimberlites, and is the first report of such serpentines from an igneous paragenesis.

ACKNOWLEGEMENTS

This study is supported by the Natural Sciences and Engineering Research Council of Canada and Lakehead University. We thank Dave Newling and Al McKenzie for assistance with the scanning electron microscopes at the University of Cambridge and at Lakehead University, respectively. Fred Wicks, Royal Ontario Museum, is thanked for assistance with the interpretation of electron-diffraction data. The referees are thanked for constructive reviews.

REFERENCES

- BATES, T.F. (1961): Electron microscopy of minerals. In The Encyclopaedia of Microscopy (G.L. Clark, ed.). Reinhold, New York.
- CRESSEY, B.A. (1979): Electron microscope studies of serpentine textures. Can. Mineral. 17, 741–756.
 - <u>& Zussman, J. (1976): Electron microscopic</u> studies of serpentinites. *Can. Mineral.* **14**, 307-313.
- JAGO, B.C. & MITCHELL, R.H. (1985): Mineralogy and petrology of the Ham kimberlite, Somerset Island, Northwest Territories. *Can. Mineral.* 23, 619-634.
- JIANG, SHAOYING, & LIU, WEI (1984): Discovery and significance of Povlen-type hydrochrysotile. Acta Geol. Sinica (Dizhi Xuebao) 58, 136-142 (in Chinese).
- KORNILOVA, V.P., NIKISHOVA, L.O. NIKISHOV, K.N. (1981): Minerals of the serpentine group from Yakutian kimberlites. *In* Mineral Parageneses in Kimberlites. Nauka Press, Yakutsk, USSR (in Russ.).

- KRSTANOVIĆ, I. & PAVLOVIĆ, S. (1964): X-ray study of chrysotile. Amer. Mineral. 49, 1769-1771.
 - _____ & _____ (1967): X-ray study of six-layer orthoserpentine. Amer. Mineral. 52, 871-876.
- LIVI, K.J.T. & VEBLEN, D.R. (1987): "Eastonite" from Easton, Pennsylvania: a mixture of phlogopite and a new form of serpentine. *Amer. Mineral.* 72, 113-125.
- LODOCHNIKOV, V.N. (1933): Serpentines and serpentinites and the petrological problems connected with them. *Prob. Soviet Geol.* 2, 145-150 (in Russ.).
- MELLINI, M. (1986): Chrysotile and polygonal sepentine from the Balangero serpentinite. *Mineral. Mag.* 50, 301-305.
- MIDDLETON, A.P. & WHITTAKER, E.J.W. (1976): The structure of Povlen-type chrysotile. *Can. Mineral.* 14, 301-306.
- MITCHELL, R.H. (1986): Kimberlites: Mineralogy, Geochemistry and Petrology. Plenum Press, New York.
- PODVYSOTKIY, V.T. (1985): Serpentine-carbonate mineralization in kimberlites. Zap. Vses. Mineral. Obshchest. 114, 234-247 (in Russ.).
- , VLADIMIROV, B.M., IVANOV, S.I. & KOTEL'NIKOV, V.P. (1987): Serpentinization of kimberlite. *Dokl. Acad. Sci. U.S.S.R. Earth Sci. Sect.* **256**, 87-91.

- SKINNER, E.M.W. & CLEMENT, C.R. (1979): Mineralogical classification of Southern African kimberlites. *In* Proc. 2nd Int. Kimberlite Conf. 1, 129-139 (F.R. Boyd & H.O.A. Meyer, eds.). Amer. Geophys. Union, Washington.
- VARLAKOV, A.S. & GURYEV, S.A. (1985): Ophite and Povlen-type chrysotile in ultrabasic rocks. *Izv. Akad. Nauk SSSR, Ser. Geol.*, 72-82 (in Russ.).
- VEBLEN, D.R. & BUSECK, P.R. (1981): Hydrous pyriboles and sheet silicates in pyroxenes and uralites: intergrowth microstructures and reaction mechanism. *Amer. Mineral.* 66, 1107-1134.
- WHITTAKER, E.J.W. & ZUSSMAN, J. (1971): The serpentine minerals. *In* The Electron-Optical Investigation of Clays (J.A. Gard, ed.). Mineral. Soc., London.
- WICKS, F.J. & WHITTAKER, E.J.W. (1977): Serpentine textures and serpentinization. *Can. Mineral.* 15, 459-488.
- diffraction patterns of the serpentine minerals. *Can. Mineral.* **13**, 244-258.
- ZUSSMAN, J., BRINDLEY, G.W. & COMER, J.J. (1957): Electron diffraction studies of serpentine minerals. *Amer. Mineral.* 42, 133-153.
- Received October 16, 1987, revised manuscript accepted February 22, 1988.