

## Seifertite, a dense orthorhombic polymorph of silica from the Martian meteorites Shergotty and Zagami

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**Abstract:** Seifertite is a dense orthorhombic polymorph of silica with the scrutinyite ( $\alpha$ -PbO<sub>2</sub>) type structure that was found as lamellae occurring together with dense silica glass lamellae in composite silica grains in the heavily shocked Martian meteorite Shergotty. The mineral is also intergrown in some grains with minor stishovite and a new unnamed monoclinic dense silica polymorph with a ZrO<sub>2</sub>-type structure. Seifertite has also been found in the Martian shergottite Zagami and is a minor constituent in other Martian shergottites. Chemical analyses of seifertite in Shergotty indicate major SiO<sub>2</sub> with minor concentrations of Al<sub>2</sub>O<sub>3</sub> and Na<sub>2</sub>O. Selected-area electron diffraction (SAED) and X-ray diffraction can be interpreted in terms of an orthorhombic pattern from a scrutinyite ( $\alpha$ -PbO<sub>2</sub>) structure. The cell parameters are  $a = 4.097(1)$  Å,  $b = 5.0462(9)$  Å,  $c = 4.4946(8)$  Å,  $V = 92.92$  Å<sup>3</sup>,  $Z = 4$ , and the space group is  $Pbcn$  or  $Pb2n$ . Density is (calc.) = 4.294 g/cm<sup>3</sup> (with pure SiO<sub>2</sub>), 4.309 g/cm<sup>3</sup> (with empirical formula). It is inferred that seifertite was formed by shock-induced solid-state transformation of either tridymite or cristobalite on Mars at an estimated minimum equilibrium shock pressure in excess of 35 GPa. The new mineral is named after Friedrich A. Seifert (b. 1941), founding Director of the Bayerisches Geoinstitut, Universität Bayreuth, Germany, for his seminal contributions to high-pressure geoscience.

**Key-words:** seifertite, new mineral, silica, high-pressure phases,  $\alpha$ -PbO<sub>2</sub> structure type, shock metamorphism, Shergotty meteorite.

### Introduction

Since the discovery of coesite and stishovite in Meteor Crater in Arizona (Chao *et al.*, 1960, 1962) and in the Ries Crater in Germany (Shoemaker & Chao, 1961; Chao & Littler, 1962), and the finding of coesite in exhumed ultra-high pressure metamorphic rocks (Chopin, 1984; Gillet *et al.*, 1984), there has been significant interest in the existence of denser silica polymorphs. There are two closely-related themes in the exploration of these higher density phases. First, whether very dense silica polymorphs exist either in the deep mantle (*e.g.* Hemley *et al.*, 1994) or at even higher pressures (Kuwayama *et al.*, 2005). Second, whether the phase transitions from low-pressure polymorphs of silica induced by dynamic compression can constrain the conditions of impact events on planetary surfaces.

Silica polymorphs denser than coesite have never been encountered in any exhumed rocks on Earth. Planetary material that has been subjected to dynamic pressures in excess of  $\geq 35$  GPa remains the best candidate for search-

ing for stishovite and other denser natural polymorphs. It has been known for many years that the basaltic shergottites, a category of SNC meteorites (Shergotty, Nakhla, Chassigny), widely considered to be of Martian origin, contain an accessory silica mineral (Tschermak, 1872, 1883; Duke, 1968). The nature of this mineral, however, has been controversial for more than four decades. It was once described as cristobalite (Duke, 1968) or as  $\alpha$ -quartz with shock-induced planar defects (Stöffler *et al.*, 1986). Shergottites also display other deformation features suggesting that they have been subjected to high-pressure conditions in a dynamic event on their parent body (Stöffler *et al.*, 1986; Müller, 1993; Chen & El Goresy, 2000; Langenhorst & Poirier, 2000a, b, c; Beck *et al.*, 2004; Malavergne *et al.*, 2001; Chennaoui-Aoudjehane *et al.*, 2005; Chennaoui-Aoudjehane & Jambon, 2006).

An orthorhombic silica polymorph denser than stishovite was found in Shergotty and characterized by Sharp *et al.* (1999), Dera *et al.* (2002) and El Goresy *et al.* (2004). Its description as a new mineral was submitted to the

Commission on New Minerals and Mineral Names, International Mineralogical Association, and approved with the mineral name seifertite. The name honours Friedrich A. Seifert (b. 1941), founding Director of the Bayerische Geoinstitut, Universität Bayreuth, Germany, for his important contributions to high-pressure geoscience. We report here this dedication and recall the main features of the new species; we also examine the  $P - T$  relevance of the silica-bearing assemblages and evaluate current information on the high-pressure assemblages in Shergotty, Zagami and some other shergottites in order to estimate the conditions of seifertite formation in the Martian meteorites.

### Occurrence and petrography, distinguishing features

Shergotty is a basaltic achondrite that fell on August 25, 1865 in Bihar State in India. The main mass of 3600 grams is preserved at the Museum of the Geological Survey in Calcutta, India. The meteorite consists of 70 % pyroxene and 24 % glass with labradorite composition or “maskelynite” (Tschermak, 1872, 1883). Minor constituents are titanomagnetite, ilmenite, pyrrhotite and silica. Silica grains, up to 900  $\mu\text{m}$  in size, are usually enclosed in maskelynite and rarely border pyroxene (Sharp *et al.*, 1999; El Goresy *et al.*, 2000, 2004; Dera *et al.*, 2002). Rare shock melted mesostasis pockets with much smaller prismatic silica grains are also encountered.

All silica grains consist of a lamellar intergrowth of at least two dense polymorphs and dense silica glass (Sharp *et al.*, 1999; El Goresy *et al.*, 2000; El Goresy *et al.*, 2004; Chennaoui-Aoudjehane *et al.*, 2005; Chennaoui-Aoudjehane & Jambon, 2006). The large seifertite-bearing silica grains show the typical pre-shock morphology and habit of tridymite or cristobalite (Sharp *et al.*, 1999; El Goresy *et al.*, 2000) but not quartz as suggested by Stöffler *et al.* (1986). Each grain is surrounded by pervasive radiating fractures that initiate at its surfaces and penetrate up to 600  $\mu\text{m}$  into the Shergotty matrix (Fig. 1A) (Sharp *et al.*, 1999; El Goresy *et al.*, 2000; Chen & El Goresy, 2000) which indicate a large volume increase due to relaxation after decompression. They thus represent a fundamental texture for *in situ* recognition of other polymorphs of silica denser than stishovite. Every silica grain consists of a mosaic of domains (10–60  $\mu\text{m}$  in size) each of which displays two orthogonal sets of lamellae that have different brightness in reflected-light microscopy. Prismatic silica grains in mesostasis show the same textural pattern. The difference in brightness or reflectance results from the differences in the refractive indices (R.I.) and densities: species with higher R.I. and density are brighter than polymorphs of lower R.I. and density. This brightness contrast is also easily recognizable in BSE mode in the Field-Emission SEM (FESEM) due to the differences in densities of the lamellar sets (Fig. 1A and 1B). Recognition of this lamellar intergrowth in reflected light and BSE in SEM is a key method in tracking the silica polymorphs denser than stishovite. Transmitted-light examination of sections of standard thickness ( $\sim 25 \mu\text{m}$ ) is hampered by blurring

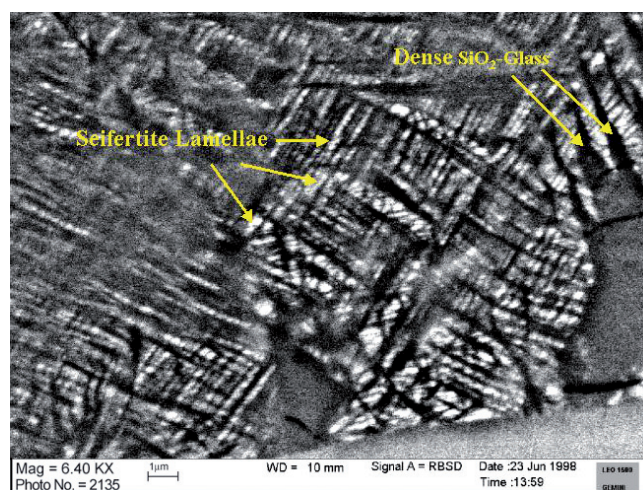
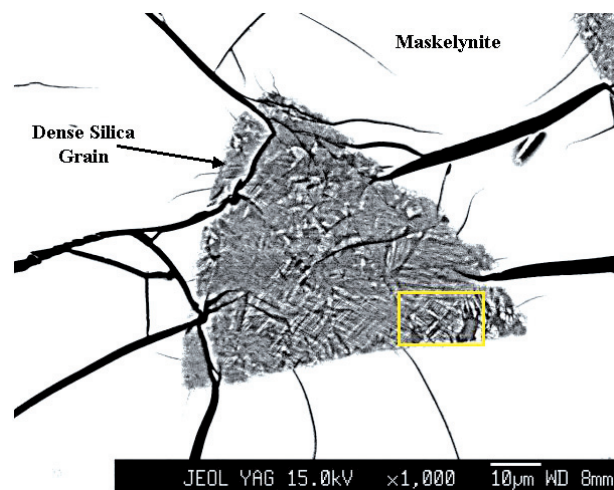


Fig. 1. (A) Backscattered electron (BSE) micrograph of a triangular dense silica grain from the Shergotty meteorite. The grain consists of numerous domains of a maximum diameter of 60  $\mu\text{m}$ . Each domain displays an orthogonal pattern of bright (seifertite) and dark (dense  $\text{SiO}_2$  glass) lamellae. Reproduced with permission from Sharp *et al.* (1999) and El Goresy *et al.* (2004). (B) BSE detail of the area within the yellow box in Fig. 1A. White lamellae are seifertite and dark to black lamellae consist of dense  $\text{SiO}_2$  glass, the latter probably formed by vitrification of another metastable dense silica polymorph of unknown nature. Difference in brightness between seifertite lamellae (high) and silica glass (low) reflects the difference in density (the denser seifertite is brighter) since both lamellae have the same chemical composition. Reproduced with permission from El Goresy *et al.* (2004).

due to the sub-micrometer lamellar intergrowths. In SEM-BSE pictures narrow, bright, and dense seifertite lamellae (usually  $\leq 200 \text{ nm}$ ) alternate with darker lamellae of less-dense silica glass of the same width and chemical composition (Fig. 1B) (Sharp *et al.*, 1999). Lamellae in silica grains in Zagami are wider (up to 1  $\mu\text{m}$ ) than those in Shergotty. This texture can be distinguished from shock-induced planar deformation lamellae in quartz or cristobalite, which exhibit much lower reflectivity in reflected light, and lower brightness than maskelynite in BSE-SEM.

We emphasize that this orthogonal texture is not restricted to seifertite-bearing grains, since it has also been

encountered in other silica grains in Shergotty that contain another new monoclinic dense polymorph plus seifertite, stishovite, as well as a secondary cristobalite-stishovite topotactic intergrowth and secondary cristobalite that are ion-milling artifacts (El Goresy *et al.*, 2000; Malavergne *et al.*, 2001). Meanwhile, silica grains with lamellar structure, similar to seifertite-bearing grains, were also encountered in numerous shergottites and were identified as such by cathodoluminescence (Chennaoui-Aoudjehane *et al.*, 2005; Chennaoui-Aoudjehane & Jambon, 2006).

## Chemistry

Electron-microprobe analyses (EMPA) with a defocused beam on the widest seifertite and glass lamellae, respectively, showed almost pure SiO<sub>2</sub> with minor concentrations in Na<sub>2</sub>O (0.2 to 0.50 wt.%) and Al<sub>2</sub>O<sub>3</sub> (0.8 to 1.60 wt.%) (Sharp *et al.*, 1999; El Goresy *et al.*, 2000; Dera *et al.*, 2002). A recent survey of many silica grains in the shergottite NWA 856 (Jambon *et al.*, 2008) revealed a wider range in the Al<sub>2</sub>O<sub>3</sub>-content (0.4–2.2 wt.%) than reported before. SIMS analyses of seifertite-bearing silica grains in both Shergotty and Zagami (Boctor *et al.*, 2003) show that they are hydrous, containing  $30 \pm 3$  to  $81 \pm 15$  ppm water. It was not possible to assess if the measured water was present only in the seifertite lamellae, in the SiO<sub>2</sub> glass lamellae, or in both. The  $\delta D$  for the analyzed spots in the silica grains ranged from  $1793 \pm 61$  to  $1246 \pm 38$  ‰ for Shergotty and  $2303 \pm 53$  to  $1173 \pm 77$  ‰ for Zagami. The fractionated H isotope signature in seifertite-bearing silica suggests partial equilibration with a highly fractionated water reservoir on Mars. Hydrogen devolatilization during impact would lead to D enrichment, though devolatilization alone could not account for the highly fractionated H isotope signature of the seifertite-bearing silica grains. The process and timing of water incorporation with respect to the shock event and decompression are still unclear.

## Crystallography

The crystal structure of seifertite was determined on two separate grains independently extracted and investigated by TEM (Sharp *et al.*, 1999) and powder X-ray diffraction (Dera *et al.*, 2002). Rietveld refinement of the latter definitively established the structure as that of scrutinyite,  $\alpha$ -PbO<sub>2</sub>, with unit-cell parameters  $a = 4.097(1)$ ,  $b = 5.0462(9)$ ,  $c = 4.4946(8)$  Å and space group  $Pbcn$ , or  $Pb2n$ . The atom coordinates are: Si in site  $4c^1$ , 0, 0.1522(9), 0.25 and O in site 8d, 0.7336(19), 0.6245(9), 0.9186(29). Figure 2 shows a polyhedral representation of the crystal structure which, like stishovite or the CaCl<sub>2</sub>-type modifications (see below), contains silicon in distorted octahedra, but with kinked chains of SiO<sub>6</sub> octahedra. The Si-O distances are 1.742 Å ( $\times 2$ ), 1.776 Å ( $\times 2$ ), 1.855 Å ( $\times 2$ ), with an average of 1.791 Å. The observed/calculated powder pattern was published by Dera *et al.* (2002).

<sup>1</sup> The z-parameter of the Si atom (0.25) was inadvertently replaced with a hyphen in Dera *et al.* (2002).

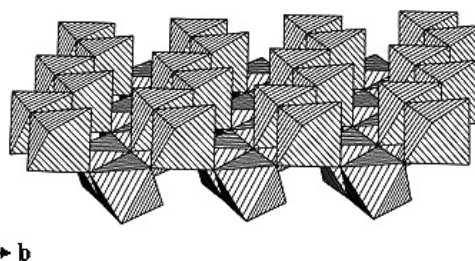


Fig. 2. Polyhedral representation of the structure of seifertite.

## Precautions for sample handling of seifertite and related dense polymorphs

Silica polymorphs denser than stishovite are very sensitive to any kind of irradiation (focused electron, laser or ion beams). They quickly amorphize (El Goresy *et al.*, 2000; Sharp *et al.*, 1999), or partially invert to a topotactic intergrowth of stishovite and cristobalite, either during standard ion-beam milling or electron irradiation in the TEM (*e.g.* El Goresy *et al.*, 2000; Malavergne *et al.*, 2001; Chennaoui-Aoudjehane *et al.*, 2005). They may also partially amorphize during prolonged X-ray diffraction under ambient laboratory conditions (El Goresy *et al.*, 2000). However multiple exposures as long as 24 h with 1.2 kW beam did not produce any damage in seifertite, nor did synchrotron IR experiments amorphize the sample. Attempts to constrain the space group via laser Second Harmonic Generation (SHG) experiments (*e.g.* Dera *et al.*, 2002) or Raman microprobe spectroscopy were not successful so far due to the fact that the post-stishovite crystalline SiO<sub>2</sub> material is unstable under the laser beam and easily vitrifies during acquisition of the Raman spectrum (Sharp *et al.*, 1999).

These problems restrict the types and sequence of the experiments for characterizing post-stishovite natural silica polymorphs to essentially the following; (1) search and documentation in reflected-light microscopy followed by cathodoluminescence studies at the SEM in scanning or TV modes only and at low beam current (El Goresy *et al.*, 2000, 2004; Chennaoui-Aoudjehane & Jambon, 2006); (2) X-ray diffraction for limited periods of time; (3) electron-microprobe analysis only with defocused beam, at low beam current, and for very short periods of time; (4) as a last technique, ion milling for TEM with low ion energy and cooling the sample with liquid N<sub>2</sub> or, better, extracting thin slices of the grains by focused-ion-beam (FIB) cutting; (5) TEM investigations at low electron dose, preferably with a liquid-N<sub>2</sub> cooling sample holder. Ignoring these precautions will not only lead to flawed results but also to irreversible damage of the meteorite sample (*e.g.* Sharp *et al.*, 1999; El Goresy *et al.*, 2000; Weber *et al.*, 2000).

## Discussion

Transitions to silica phases denser than stishovite have been identified in both first-principles calculations (*e.g.* Cohen, 1992; Teter *et al.*, 1998; Karki *et al.*, 1997; Belonoshko

*et al.*, 1996) and experiments (*e.g.* Hemley *et al.*, 1994; Kingma *et al.*, 1995; Dubrovinsky *et al.*, 1997). The equilibrium phase transition sequence with increasing pressure is stishovite (rutile-type) – CaCl<sub>2</sub>-type –  $\alpha$ -PbO<sub>2</sub>-type (Teter *et al.*, 1998), and both phase boundaries have positive *P-T* slopes (*e.g.* Murakami *et al.*, 2003). Comparison of the results of Dubrovinsky *et al.* (1997, 2001) with those obtained by Murakami *et al.* (2003) also suggests that  $\alpha$ -PbO<sub>2</sub>-type silica could be synthesized well outside the inferred stability field of the phase. This is confirmed by the synthesis of  $\alpha$ -PbO<sub>2</sub>-type species (space group *Pbcn* or *Pb2n*) at  $P \geq 40$  GPa and 300 K (Tsushida & Yagi, 1990; Dubrovinsky *et al.*, 2001; Dubrovinskaja *et al.*, 2001) from natural cristobalite.

The estimation of the minimum equilibrium pressure required to induce the inversion of the parental low-pressure silica polymorph to seifertite can be improved on the basis of recent reports of a large number of shock-induced high-pressure minerals in various shergottites.

Shergotty contains two distinct high-pressure assemblages in different settings: (1) seifertite-bearing maskelynite grains, and (2) shock-melt pockets that are barren of any seifertite-bearing silica grains but consist of lingunite (hollandite-structured labradorite composition) (*cf.* Chen *et al.*, 2000), a new calcium hexaluminosilicate (CAS) (Irifune *et al.*, 1994; Gautron *et al.*, 1999; Beck *et al.*, 2004), and large individual stishovite grains up to several tens of micrometers in diameter that show no tweed pattern (Beck *et al.*, 2003, 2004; El Goresy *et al.*, 2004). In this meteorite the high-pressure associations in pockets are confined to seifertite-free assemblages. Accordingly, these pockets crystallized below the lower-pressure stability bound of seifertite. Beck *et al.* (2004) interpret the seifertite-free assemblage in shock-melt pockets as indicative of pressures clearly in excess of 25 GPa and temperatures between 2300 and 2500 °C.

Zagami contains in addition *shock-melt veins* with a variety of dense minerals consisting of omphacite, acicular stishovite, KAlSi<sub>3</sub>O<sub>8</sub> hollandite, Na-, Ca-rich hollandite (lingunite) in addition to akimotoite, amorphized perovskite and silicate titanite (Langenhorst & Poirier, 2000a, b; Chen *et al.*, 2000) but no CAS or Ca-ferrite structured NaAlSiO<sub>4</sub> (Yagi *et al.*, 1994). The high-pressure mineral inventory of the veins varies from the vein centers to the rims depending on the minerals in contact with them. Langenhorst & Poirier (2000a, b) reasoned that the polycrystalline hollandite aggregates in the center of veins with plagioclase from the neighboring matrix reworked in the silicate melt record pressures < 23 GPa, whereas pyroxene observed in the vein margins may have formed at < 10 GPa. This conclusion is consistent with the interpretation that the vein high-pressure assemblages crystallized in the decompression stage. Such low pressures at the vein margin are insufficient to induce the tridymite (or cristobalite)-to-seifertite phase transition in the meteorite matrices and indicate that the local process that led to the vein formation is unrelated to the tridymite (or cristobalite)-to-seifertite phase transition in the matrix (*cf.* Xie *et al.*, 2005).

As in Shergotty, *shock-melt pockets* in Zagami contain, in addition to hollandite-structured labradorite composi-

tion, the high-pressure liquidus pair acicular stishovite + CAS (Fig. 4 and 5 in Beck *et al.*, 2004) and no seifertite-bearing silica grains or Ca-ferrite structured NaAlSiO<sub>4</sub>. The pockets both in Shergotty and Zagami do not show any variation in the high-pressure inventory from pockets centers to their rims to the matrix. The mineral record in the Zagami pockets also indicates formation at pressures and temperatures like their counterparts in Shergotty (Beck *et al.*, 2004). In short, we consider the absence of seifertite-bearing silica grains in pockets and veins in these two shergottites a strong evidence that the tridymite (or cristobalite)-to-seifertite phase transitions in the meteorite matrices required higher equilibrium pressures than the upper bound set for veins (23 to 10 GPa, Langenhorst & Poirier, 2000a, b) and pockets (25 GPa, Beck *et al.*, 2004). Lakshtanov *et al.* (2007) demonstrate that the required pressure for stishovite/CaCl<sub>2</sub>-polymorph inversion decreases with the increase of the Al<sub>2</sub>O<sub>3</sub>-content of the parental phase. This result may suggest that the Al<sub>2</sub>O<sub>3</sub>-contents of the individual parental low-pressure polymorphs may have a considerable influence on the required pressures for the phase transitions of every individual parental grain to seifertite and/or stishovite.

The inventory of high-pressure phases in shock melt pockets and veins in Shergotty and Zagami meteorites suggests that the parental tridymite (or cristobalite)-to-seifertite phase transformation was induced in a shock event on Mars at pressures probably in excess of 35 GPa (Langenhorst & Poirier, 2000a, b; Beck *et al.*, 2004), with the caveat that the *P-T* slope of the metastable tridymite (or cristobalite)-seifertite phase boundary is not well defined. Therefore, this estimate of the equilibrium shock pressure is only a lower bound. The transformation resulted in seifertite plus another unknown metastable dense polymorph, the latter amorphized to dense silica glass on decompression. No residues of the parental low-pressure polymorph were encountered in the investigated grains, indicating that the reconstructive phase transition was indeed complete.

**Acknowledgements:** The senior author expresses his gratitude to Gero Kurat, Naturhistorisches Museum, Vienna, and Monika Grady, British Museum, London, for the Shergotty samples provided. The disc studied by Xie *et al.* was cored out a Zagami polished thin section from the collection of the senior author. Przemyslaw Dera and his colleagues at the Geophysical Laboratory, Carnegie Institution of Washington, are grateful to Timothy McCoy, Smithsonian Institution, Washington D.C., for providing the Shergotty sample from which they extracted the seifertite grain for X-ray studies. The authors are grateful to an anonymous reviewer, Managing Editor Christian Chopin, Associate Editor Ross Angel and Chief Editor Roland Oberhänsli for their suggestions and comments, which helped in improving the manuscript. This work was in part supported by NSF (EAR) and DOE (CDAC).

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Received 16 November 2007

Modified version received 7 February 2008

Accepted 25 February 2008