



Article

Chrysoberyl and associated beryllium minerals resulting from metamorphic overprinting of the Maršíkov–Schinderhübel III pegmatite, Czech Republic

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Abstract

The Maršíkov–Schinderhübel III pegmatite in the Hrubý Jeseník Mountains, Silesian Domain, Czech Republic, is a classic example of chrysoberyl-bearing LCT granitic pegmatite of beryl–columbite subtype. This thin pegmatite dyke, (up to 1 m in thickness in biotite–amphibole gneiss is characterised by symmetrical internal zoning. Tabular and prismatic chrysoberyl crystals (\leq 3 cm) occur typically in the intermediate albite-rich unit and rarely in the quartz core. Chrysoberyl microtextures are quite complex; their crystals are irregularly patchy, concentric or fine oscillatory zoned with large variations in Fe content (1.1–5.3 wt.% Fe₂O₃; \leq 0.09 apfu). Chrysoberyl compositions reveal dominant Fe³⁺ = Al³⁺ and minor Fe²⁺ + Ti⁴⁺ = 2(Al, Fe)³⁺ substitution mechanisms in the octahedral sites. Tin, Ga, and V (determined by LA-ICP-MS) are characteristic trace elements incorporated in the chrysoberyl structure, whereas anomalously high Ta and Nb concentrations (thousands ppm) in chrysoberyl are probably caused by nano- to micro-inclusions of Nb–Ta oxide minerals; especially columbite–tantalite. Textural relationships between associated minerals, distinct schistosity of the pegmatite parallel to the host gneiss foliation and fragmentation of the pegmatite body into blocks as a result of superimposed stress are clear evidence for deformation and metamorphic overprinting of the pegmatite. Primary magmatic beryl, albite and muscovite were transformed to chrysoberyl, fibrolitic sillimanite, secondary quartz and muscovite during a high-temperature (\sim 600°C) and medium-pressure (\sim 250–500 MPa) prograde metamorphic stage under amphibolite-facies conditions. A subsequent retrograde, low-temperature (\sim 200–500°C) and pressure (\leq 250 MPa) metamorphic stage resulted in the local alteration of chrysoberyl to secondary Fe,Na-rich beryl, euclase, bertrandite and late muscovite.

Keywords: chrysoberyl, beryl, euclase, bertrandite, sillimanite, granitic pegmatite, metamorphic overprint

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Introduction

Chrysoberyl, ideally BeAl₂O₄, is an oxide mineral of olivine-type structure with Be²⁺ cations in tetrahedral and Al³⁺ in octahedral coordination. The main substituents of Al³⁺ are Cr³⁺ (mariinskite BeCr₂O₄, a Cr-dominant member; Pautov *et al.*, 2013) and Fe³⁺ (up to ~6 wt.% Fe₂O₃, Žáček and Vrána, 2002). Alexandrite, a gemstone variety of chrysoberyl, is characterised by two distinct optical effects, including the alexandrite effect produced by Cr³⁺ and V³⁺ impurities and the cat's eye effect. Three principal lithological types of magmatic or metamorphic chrysoberyl occurrences have been recognised: (1) granitic pegmatites and exceptionally leucogranites; (2) metasomatic skarn zones and related dykes

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along/near the contacts between ultrabasic or carbonate rocks and intruded granites to pegmatites; and (3) high-temperature metamorphic rocks of granulite facies (e.g. Martin-Izard *et al.*, 1995; Barton and Young, 2002; Downes and Bevans, 2002; Černý, 2002; Franz and Morteani, 2002; Marschall and Walton, 2014).

Chrysoberyl has been described as an accessory mineral in numerous rare-element, peraluminous granitic pegmatites of the beryl type or, rarely, in pegmatites of the abyssal class (Grew, 1981; Černý and Ercit, 2005; Cempírek and Novák, 2006). Chrysoberyl is usually associated with primary rock-forming minerals such as quartz, feldspars and muscovite, and locally with beryl. However, the origin of chrysoberyl in granitic pegmatites is typically uncertain: it is considered to be a product of primary magmatic precipitation or metamorphic overprinting of a parental pegmatite. A primary magmatic origin for chrysoberyl is suggested for pegmatites in Southern Kerala, India (Soman and Nair, 1985), the Malga Garbella pegmatite, Italy (Vignola *et al.*, 2018), Tablada I pegmatite, Argentina (Colombo *et al.*, 2021) and the Tashisayi pegmatites,

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China (Hong et al., 2021). Magmatic chrysoberyl in assemblage with beryl, sillimanite and gahnite was described from the highly-evolved, peraluminous Belvís de Monroy leucogranite, Spain (Merino et al., 2013). However, most occurrences of chrysoberyl consider it to have formed from the breakdown of primary beryl as a result of metamorphic overprinting of the pegmatite e.g. Kolsva, Sweden; Haddam, USA; and Maršíkov, Czech Republic (Franz and Morteani, 1984, 2002; Černý et al., 1992); Kalanga Hill, Zambia (Žáček and Vrána, 2002); Ethel Mary and Virorco, Argentina (Lira and Sfragulla, 2011; Galliski et al., 2012); New York, USA (Lupulescu et al., 2012); Roncadeira, Brazil (Beurlen et al., 2013); and Mt. Begbie, Canada (Dixon et al., 2014).

In this investigation we examine the composition of chrysoberyl and associated minerals at the Maršíkov–Schinderhübel III granitic pegmatite, the first reported occurrence of chrysoberyl in Europe (Hruschka, 1824). The principal objectives of our study were to characterise textural, paragenetic and compositional relations of chrysoberyl and associated Be minerals, and to estimate the conditions of formation of assemblages containing these minerals, and to relate these to the metamorphic evolution of the pegmatites.

Regional geology and pegmatite description

The granitic pegmatite occurs in the Desná Dome, Neoproterozoic to Devonian crystalline basement of the Silesian Domain of Bohemian Massif, a part of the European Variscan orogenic belt (Fig. 1). The Silesian Domain represents an orogenic wedge belonging to the Moravo–Silesian Zone, a metamorphosed and imbricated margin at the eastern Variscan front in NE Bohemian Massif (Schulmann and Gayer, 2000; Schulmann *et al.*, 2014). The central part of the Desná Dome consists of orthogneisses, paragneisses and amphibolites showing Neoproterozoic protolith ages (570–650 Ma; Kröner *et al.*, 2000; Jastrzębski *et al.*, 2021) and Palaeozoic cover rocks, mainly metaquartzites, metaconglomerates, metapelites and marbles, which are intruded locally by Middle Devonian (Givetian) volcanic rocks of arc and back-arc affinity (Janoušek *et al.*, 2014). This metamorphic complex underwent Variscan medium-pressure, Barrovian-type metamorphism at $P \approx 500$ –700 MPa and $T \approx 540$ –660°C (Souček, 1978; René, 1983, Cháb *et al.*, 1990; Schulmann and Gayer, 2000; Košuličová and Štípská, 2007; Schulmann *et al.*, 2014).

The metamorphic rocks were intruded by Variscan, Carboniferous to Early Permian granitic rocks and pegmatites. Zircon U–Pb radiometric age determination of pegmatitic leucogranite at Čertovy Kameny hill near Jeseník gave 334 Ma (Hegner and Kröner, 2000), which is consistent with ages of older granitic plutons in the adjacent Sudetic block of NE margin of the Bohemian Massif: Kłodzko-Złoty Stok, Jawornik, Strzelin, and Kudowa plutons (350–330 Ma; Mikulski *et al.*, 2013). Moreover, a younger plutonic suite of I-type granodiorites to granites of the Žulová Pluton have late-Variscan, Early Permian emplacement ages of 292 ± 4 or 291 ± 5 Ma (LA–ICP–MS U–Pb zircon radiometric age determination; Laurent *et al.*, 2014).

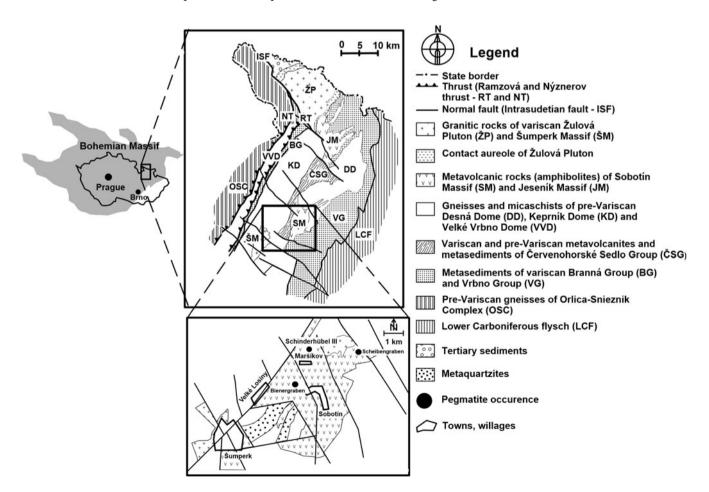


Figure 1. Simplified geological situation of the Maršíkov District with individual pegmatite occurrences (Cháb et al., 2007 and on-line map application of the Czech Geological Survey 2019, Chládek et al., 2020, modified).

The metamorphic rocks of the Desná Dome were intruded primarily in the Maršíkov pegmatite district by numerous dykes or lenticular bodies of granitic pegmatites enclosed in biotite-amphibole gneisses and/or amphibolites of the Sobotín Massif (Fig. 1). The most fractionated pegmatites show an affinity to the LCT (lithium-caesium-tantalum) family and beryl-columbite subtype of the rare-element class according to the classification of Černý and Ercit (2005), as were documented by their mineral assemblages (e.g. Černý et al., 1992, 1995; Novák 1988, 2005; Novák et al., 2003, 2023; Chládek and Zimák, 2016; Chládek et al., 2020, 2021; Dolníček et al., 2020a, 2020b).

The Maršíkov-Schinderhübel III chrysoberyl-bearing granitic pegmatite is situated on the hill NNE of the Maršíkov settlement, a part of Velké Losiny village, ~10 km NE from Šumperk town in the southern part of the Hrubý Jeseník Mountains, northern Moravia region, Czech Republic. It forms a dyke in biotite-hornblende gneiss belonging to Sobotín amphibolite massif. The pegmatite is not exposed, excavation trenches show it to be up to 1 m thickness, with a 160° NNW-SSE strike and inclination of 20° to SW, concordantly to the adjacent gneiss (Dostál, 1966). The pegmatite dyke shows symmetrical internal zoning, including an aplitic unit, an intermediate albite-rich unit with coarse-grained muscovite, and a quartz core unit (Dostál, 1966; Staněk, 1981; Černý et al., 1992; Fig. 2). The aplitic unit is only ~1 cm in thickness and consists of a fine-grained (~0.5 mm) aggregate of albite, quartz, muscovite and garnet (almandine-spessartine) in subordinate amounts, and rare biotite. The intermediate albite-rich unit, ~15-50 cm in thickness, is composed of a medium- to coarse-grained aggregate of albite, quartz and muscovite with abundant fibrolitic sillimanite and relatively common chrysoberyl, garnet, fluorapatite, gahnite, Nb-Ta oxide minerals (columbite- and microlite-group members and fersmite), zircon, uraninite, cheralite, native bismuth and bismutite. Sillimanite, chrysoberyl and muscovite of this unit exhibit a distinct

metamorphic grain lineation parallel to foliation of the pegmatite. The central quartz core unit is ~10 to 40 cm in thickness, and is characterised by less-developed schistosity and contains aggregates of anhedral quartz in association with muscovite, beryl, rare chrysoberyl (more common along the contact of the quartz core with the intermediate unit), sillimanite, garnet, gahnite, and Nb–Ta minerals. The entire Schinderhübel III pegmatite is deficient in K-feldspar (Dostál, 1966; Černý et al., 1992). The other smaller pegmatite dykes with Be minerals (Schinderhübel I and II) occur ~80 m east and ~50 m northwest of the Schinderhübel III pegmatite, and both exhibit similar mineral assemblages with minor to rare K-feldspar and noticeable deformation although accompanied by a lesser degree of metamorphic overprinting and paucity of chrysoberyl and sillimanite (Staněk, 1981; Černý et al., 1992).

Methods

The samples investigated were obtained from several sources, including the Moravian Museum, Brno mineralogical collection and the research collections of S. Chládek and M. Novák. The abbreviations for the minerals used in the text and figures are after Warr (2021). The composition of minerals were determined on polished sections using a JEOL JXA-8530F field-emission electron-probe microanalyser (EPMA) using wavelength dispersive spectrometry (WDS) at the Institute of the Earth Sciences of the Slovak Academy of Sciences in Banská Bystrica, Slovakia. The following analytical conditions were used: accelerating voltage 15 kV; probe current 20 nA; beam diameter ranging from 3 to 5 µm; and the ZAF matrix correction. The following standards and X-ray lines were used: diopside (SiKα, MgKα, CaKα); rutile (TiKα); albite (AlKα, NaKα); cassiterite (SnLα); ScVO₄ (ScK α , VK α); Cr₂O₃ (CrK α); GaAs (GaL α); YPO₄ (YL α); hematite (FeK α); rhodonite (MnK α); forsterite (MgK α); gahnite

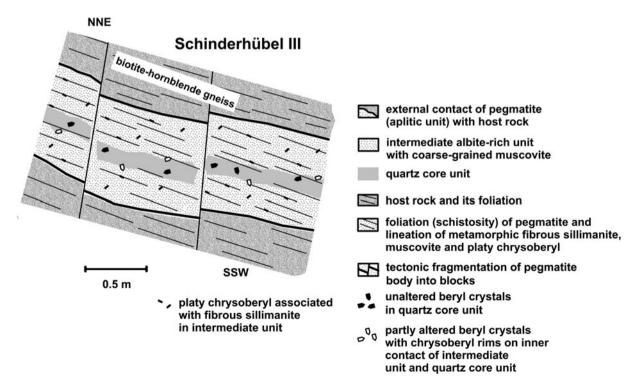


Figure 2. Idealised cross-section of the Maršíkov-Schinderhübel III pegmatite.

(ZnKα); fluorapatite (CaKα); celestine (SrLα); orthoclase (KKα); Rb₂ZnSi₅O₁₂ glass Rb (RbLα); pollucite (CsLα); and fluorite (FKα). Back-scattered electron (BSE) images and X-ray element maps were obtained on the same JEOL JXA-8530F instrument for detailed study of textural relationships among minerals. The element-distribution maps were produced using an accelerating voltage of 15 kV, probe current of 50 nA and 1 μm beam diameter. The maps comprise 1070×770 pixels, with a pixel size of 0.7 μm and a dwell time of 50 ms for each pixel.

The trace-element content of chrysoberyl was obtained by laser ablation-inductively coupled plasma-mass spectrometry (LA-ICP-MS) at the Department of Chemistry, Masaryk University, Brno, Czech Republic. The LA-ICP-MS setup consists of the laser ablation system Analyte G2+ (Teledyne Cetac) and the quadrupole ICP-MS Agilent 7900 (Agilent Technologies). The laser ablation system uses a Q-switched Nd-YAG laserablation device at the fifth harmonic frequency corresponding to 213 nm wavelength. The ablation cell was flushed with a He carrier gas, which transported the laser-induced aerosol to the inductively coupled plasma (1 l/min). Sample Ar gas flow was admixed with the He gas flow to 1.6 l/min total gas flow. ICP-MS parameters were optimised with respect to maximum signal-to-noise ratio and minimum oxide formation (ThO+/Th+ count ratio = 0.2%, U⁺/Th⁺ count ratio = 1.1%). The following laser ablation parameters were used for the analysis: 100 µm laser spot diameter; 8 J cm⁻² laser fluence; and 20 Hz repetition rate. Fixing the sample position during laser ablation facilitated a hole-drilling duration of 60 s for each spot. The NIST SRM 610 silicate glass was used as reference material. All element measurements were normalised to the average concentration of Al in the chrysoberyl, determined by the electron microprobe.

Micro-Raman analyses of minerals were performed on the LabRAM-HR Evolution Horiba Jobin-Yvon spectrometer system with a Peltier-cooled CCD detector and Olympus BX-41 microscope (Department of Geological Sciences, Masaryk University, Brno, Czech Republic). Raman spectra were excited by a blue diode laser (473 nm) with a power of 2.5 mW in the range of 100-4,000 cm⁻¹ and 100-10,000 cm⁻¹ were collected from each mineral using a 50× objective. The acquisition time of 30 s and 5 s per frame, respectively, and two accumulations were used to improve the signal-to-noise ratio. Raman spectra were also obtained using a Thermo Scientific DXR3xi Raman imaging microscope at the Natural History Museum in Bratislava, Slovakia. The doubled Nd: YVO4 DPSS excitation laser (532 nm) and He-Ne laser (633 nm), 100× objective, a 25 μm confocal pinhole, and an EMCCD detector were used. The spectra were acquired at a laser power of 10-20 mW and 0.5-2 s (20 scans for a cycle), and they were processed in the Seasolve *PeakFit 4.1.12* software. Raman bands were fitted by the Lorentz function with automatic background correction and Savitzky-Golay smoothing.

Powder X-ray diffraction (XRD) analysis was determined by the BRUKER D8 Advance diffractometer (Department of Mineralogy and Petrology, Comenius University, Bratislava) under the following conditions: Bragg–Brentano geometry (θ –2 θ), Cu anticathode ($K\alpha_1$ = 1.5406 Å), accelerating voltage = 40 kV and beam current = 40 mA; a Ni K_{β} filter was used for stripping $K\beta$ radiation, and data were obtained by a BRUKER LynxEye detector. The step size was 0.01°2 θ with a counting time of 5 s per step, and 2 θ measurement ranged from 4 to 65°. The lattice parameters were refined with the Bruker DIFFRAC plus *TOPAS* software using the structural model for chrysoberyl (Hazen and Finger, 1987).

Results

Textural and paragenetic relationships of Be minerals

The following Be minerals were identified in the pegmatite: chrysoberyl; beryl; euclase; and bertrandite. Chrysoberyl occurs in intermediate and quartz core units as transparent to translucent subhedral to anhedral rarely euhedral prismatic to tabular crystals and V-shaped twins (~50 µm to 3 cm in size) with yellowgreen to pale-green colour and vitreous to adamantine lustre. Chrysoberyl is associated with beryl, quartz, albite, muscovite and sillimanite in the intermediate unit or in its contact with the quartz core. In the outer parts of the intermediate unit, platy crystals of chrysoberyl are commonly parallel to flakes of muscovite and fibrolitic sillimanite aggregates and relics of primary beryl are absent or very rare. The BSE images of chrysoberyl exhibit mostly patchy, rarely irregular lamellar or concentric zonation of crystals (Figs 3,4,5), some large platy chrysoberyl crystals without relics of beryl show fine lamellar to oscillatory growth zoning (Fig. 3e,f). Relics of primary beryl and inclusions of quartz, albite, muscovite, gahnite, and Nb-Ta oxide minerals are commonly detected in chrysoberyl (Figs 3,4,5). Secondary Fe-rich beryl, euclase, bertrandite and late muscovite are locally confined to the edges and fractures of chrysoberyl (Figs 4b,e, 5).

Primary beryl occurs as euhedral to subhedral pale yellow-green or bluish green translucent prismatic crystals, up to 5 cm in length and 2 cm in thickness, typically in the quartz core. In the intermediate unit, crystals of primary beryl are replaced by chrysoberyl, and locally, anhedral relicts of beryl (20–300 μ m across) are associated with quartz at inner contact of the intermediate unit and quartz core units (Figs 3a,b,d, 5). Larger subhedral crystals of beryl (\sim 200 to 500 μ m) also occur with chrysoberyl and albite (Fig. 4c).

Anhedral domains of secondary Fe-rich beryl, $\sim 50 \, \mu m$ large, were rarely identified in rims of chrysoberyl crystals (Fig. 5a–d). Euclase forms subhedral to anhedral overgrowths and veinlets replacing chrysoberyl, usually $\sim 50 \, \mu m$ to 1 mm in size (Figs 3a, 4b,e, 5). Bertrandite occurs as anhedral grains or veinlets, up to 200 μm in size, located in chrysoberyl, and locally replaces euclase (Fig. 4b).

Composition of Be minerals

The zoning in BSE images (Figs 3–5) generally reflects different Fe concentrations in chrysoberyl with the lighter zones illustrating domains with elevated contents of Fe in chrysoberyl (Table 1). Valence calculations indicate the dominancy of trivalent Fe in chrysoberyl (1.1–5.3 wt.% Fe₂O₃; \leq 0.09 atoms per formula unit Fe³⁺), however commonly a small amount of Fe²⁺ (\leq 0.5 wt.% FeO) is also present, which correlates positively with Ti (\leq 0.6 wt.% TiO₂; \leq 0.01 apfu) and Sn (\leq 0.5 wt.% SnO₂; \leq 0.004 apfu). Compositional relationships suggest two main octahedral-site substitutions in chrysoberyl: (1) dominant Fe³⁺ = Al³⁺; and (2) minor Fe²⁺ + Ti⁴⁺ = 2(Al,Fe)³⁺ substitutions (Fig. 6).

The LA-ICP-MS investigation of chrysoberyl yielded elevated concentrations of Ga (\sim 240–420 ppm), Sn (\sim 40–440 ppm) and V (\sim 20–180 ppm) together with Fe and Ti (Supplementary Table S1); Cr is frequently below the detection limit. Extraordinarily high concentrations of Ta (\leq 7200 ppm) and Nb (\leq 2700 ppm) were detected locally in chrysoberyl, and they can be explained by micro- to nano-inclusions of Nb–Ta oxide minerals as documented in BSE images (Figs 3a, 4d–f, 5). Positive correlations were observed between some trace elements in chrysoberyl: Sn versus Ti, Hf vs. Zr, V vs. Ti, V vs. Ga and Ti vs. Ta (Fig. 7a–e).

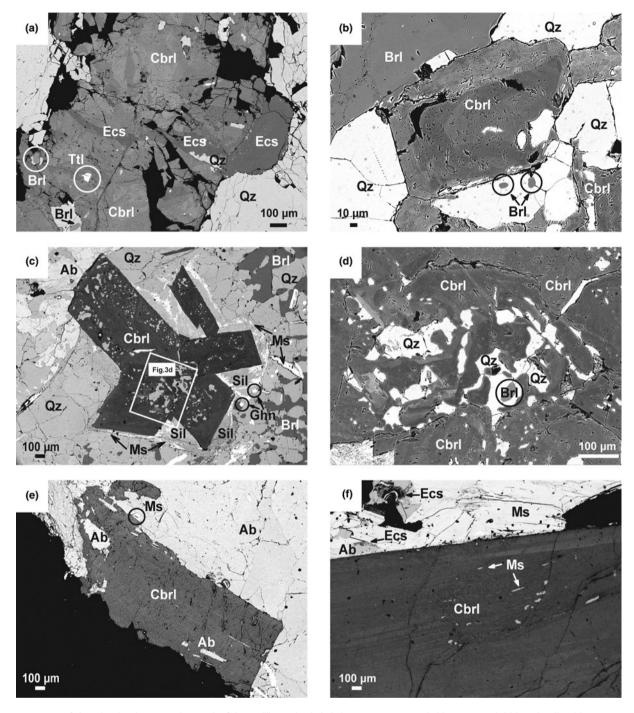


Figure 3. BSE images of chrysoberyl and associated minerals of the Maršíkov–Schinderhübel III pegmatite. Mineral abbreviations: Cbrl (chrysoberyl), Brl (primary beryl), Ecs (euclase), Qz (quartz), Ab (albite), Ms (muscovite), Sil (sillimanite), Ttl (Nb–Ta oxide minerals: tantalite, columbite and ixiolite), Ghn (gahnite).

Primary beryl is characterised by a low content of Fe: up to 1.2 wt.%; Fe $_2$ O $_3$ (\leq 0.08 apfu Fe $^{3+}$) and up to 0.5 wt.% FeO (\leq 0.04 apfu Fe $^{2+}$). Rare secondary Fe-rich beryl contains 0.7–1.1 wt.% Fe $_2$ O $_3$ (0.05–0.08 apfu Fe $^{3+}$), although it is distinctly enriched in FeO (3.1–3.4 wt.%; 0.24–0.27 apfu Fe $^{2+}$) (Fig. 5d; Table 2). Divalent Fe is compensated by Na (up to 1.6 wt.% Na $_2$ O; \leq 0.29 apfu Na) according to the channel (C) – octahedral (O) substitution: C Na $^+$ + O M(Fe,Mn,Mg,Zn) $^{2+}$ = C \Box + O Al $^{3+}$, and Fe $^{3+}$ replaces Al in octahedral, stoppaniite-type substitution: C Fe $^{3+}$ = C Al $^{3+}$ (Fig. 8). The concentrations of other elements are negligible except for 0.2 wt.% MnO (0.02 apfu Mn) in Fe-rich beryl (Table 2).

Euclase contains up to 0.4 wt.% Fe_2O_3 (0.01 apfu Fe) and negligible amounts of Zn and Ca (\leq 0.1 oxide wt.%) (Table 3). Bertrandite is also close to the end-member composition (Table 4).

Unit-cell parameters and Raman spectroscopy of the Be minerals

The unit-cell parameters of the chrysoberyl determined by powder X-ray diffraction (Table 5) are in excellent accordance with the previously published data (Dostál, 1969; Hazen and Finger, 1987). The presence of bertrandite was also confirmed by micro-Raman spectroscopy as EPMA cannot distinguish

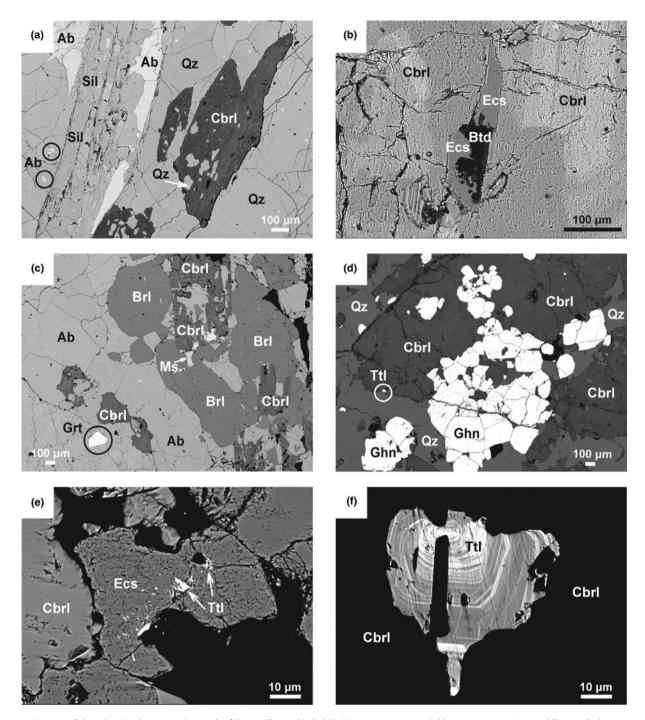


Figure 4. BSE images of chrysoberyl and associated minerals of the Maršíkov–Schinderhübel III pegmatite. Mineral abbreviations as in Fig. 3, Btd (bertrandite), Grt (garnet).

bertrandite from phenakite. The Raman spectra of bertrandite from the Maršíkov–Schinderhübel III pegmatite (Fig. 9) are in excellent agreement with the RRUFF database (R060803.3 and R050032.3 samples; Lafuente *et al.*, 2016). Note that the bertrandite spectrum also shows some typical bands for closely-associated euclase.

Discussion

Chrysoberyl composition

Several studies have focused on the description of minerals and geology from the Maršíkov-Schinderhübel III pegmatite (e.g.

Kretschmer, 1911; Dostál, 1966, 1969; Černý *et al.*, 1992); however, a detailed study of the textural relations and compositional variations in chrysoberyl and its mineral assemblage is lacking. The composition of natural chrysoberyl is typically close to the BeAl_2O_4 end-member formula. The olivine-type structure of chrysoberyl is composed of two principal cation sites: octahedrally coordinated M1 and M2 sites are occupied by Al^{3+} , and the tetrahedral (T) position is filled by Be^{2+} cations (Hawthorne and Huminicki, 2002). Aluminium in the octahedral sites is commonly replaced by other trivalent cations, especially by Cr^{3+} and Fe^{3+} . The maximum Fe_2O_3 content reported in natural chrysoberyl is 6.25 wt.% for samples from the Kalanga Hill

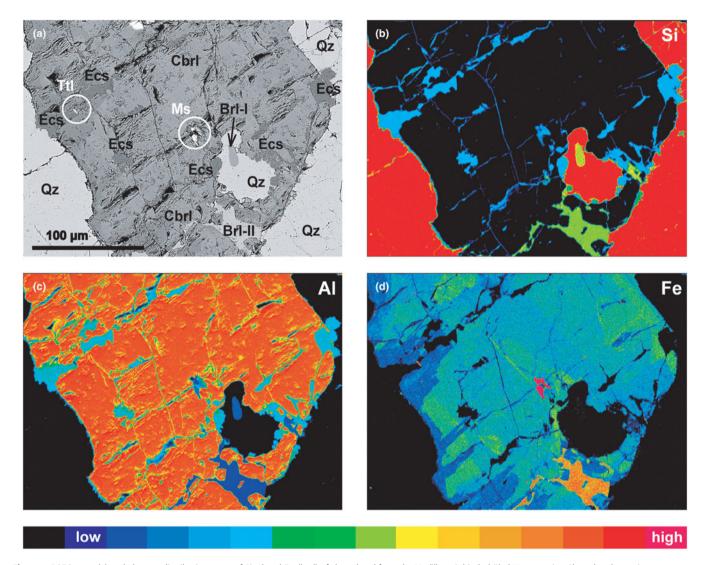


Figure 5. BSE image: (a) and element-distribution maps of Si, Al and Fe; (b–d) of chrysoberyl from the Maršíkov–Schinderhübel III pegmatite. Chrysoberyl contains tantalite inclusion, remnants of primary beryl (Brl-I), and secondary Fe-rich beryl (Brl-II), secondary muscovite and euclase. Mineral abbreviations as in Fig. 3.

pegmatite, Zambia (Žáček and Vrána, 2002). Chrysoberyl from the Maršíkov–Schinderhübel III pegmatite exhibits relatively broad variations of Fe (1.1 to 5.3 wt.% Fe_2O_3 and ≤ 0.5 wt.% FeO). Elevated Fe contents are typical of chrysoberyl of metamorphic origin, whereas magmatic chrysoberyl from leucogranites and pegmatites usually have a lower Fe content (≤ 1.2 wt.% FeO total; Merino *et al.*, 2013).

Contents of Ti (≤0.6 wt.% TiO₂; ≤0.01 apfu), Ga (~240–420 ppm), Sn (~40–440 ppm), and V (~20–180 ppm) in the Maršíkov chrysoberyl are comparable with the published data (≤0.7 wt.% TiO₂, ≤4500 ppm Sn, ≤1600 ppm Ga and (1200 ppm V; Soman and Nair, 1985; Merino *et al.*, 2013; Kanouo *et al.*, 2016; Schmetzer *et al.*, 2016; Sun *et al.*, 2019; Colombo *et al.*, 2021). High concentrations of Ta (up to ~7200 ppm) and Nb (up to ~2700 ppm), detected in the Maršíkov chrysoberyl, are very unusual and probably result from the presence of numerous nano- to micro-inclusions of Nb–Ta oxide minerals (columbite-(Fe), columbite-(Mn), tantalite-(Fe), tapiolite-(Fe), ixiolite-(Mn²⁺); Figs 3–5). Such high contents of Ta and Nb have not been found previously in chrysoberyl; ~510 ppm Ta and ~250 ppm Nb are

the maximum reported (Merino et al., 2013; Kanouo et al., 2016; Schmetzer et al., 2016; Colombo et al., 2021).

Evolution of the chrysoberyl assemblage

The origin of chrysoberyl in granitic pegmatites can be explained by two principal hypotheses: either primary magmatic or metamorphic genesis. The main criteria for distinguishing between these hypotheses are textural relationships, mineral paragenesis and the presence or absence of metamorphic overprint in the parental pegmatite. Moreover, Fe content in chrysoberyl can also be an indicator of magmatic *versus* metamorphic origin (Merino *et al.*, 2013).

Schistosity and fragmentation of the pegmatite body into blocks as a result of superimposed stress, mineral lineation and presence of fine-crystalline bands of fibrolitic sillimanite parallel to the host gneiss foliation, and replacement of beryl by chrysoberyl are distinct features of metamorphic overprinting of the Maršíkov–Schinderhübel III pegmatite (Dostál, 1966, 1969;

Table 1. Representative compositions (wt.%) and formulae (apfu) of chrysoberyl from the Maršíkov pegmatite.

Analyse #	SCH-8-1	SCH-8-3	SCH-8-4	SCH-8-5	SCH-1C-2	SCH-1C-7	SCH-2-1	SCH-2-2
SiO ₂	bdl	bdl	bdl	0.07	bdl	bdl	bdl	bdl
TiO ₂	bdl	0.08	0.62	0.18	0.11	0.08	0.10	0.12
SnO ₂	bdl	0.06	0.22	0.47	0.06	0.05	bdl	bdl
Al_2O_3	77.47	77.00	75.93	73.92	77.97	77.35	78.48	78.45
Sc_2O_3	0.03	bdl	bdl	bdl	0.06	bdl	0.03	bdl
V_2O_3	bdl	bdl	0.04	0.07	0.05	bdl	bdl	bdl
Ga_2O_3	bdl	0.07	bdl	bdl	bdl	0.12	0.08	0.04
Fe ₂ O ₃	1.98	2.96	3.79	5.29	1.69	2.36	1.27	1.11
FeO	bdl	0.06	0.53	0.09	0.13	0.04	bdl	0.07
BeO (calculated)	19.32	19.39	19.43	19.05	19.45	19.39	19.49	19.47
Total	98.80	99.62	100.56	99.14	99.52	99.39	99.45	99.26
Mineral formulae base	ed on 4 oxygen at	toms, 1 Be atom,	3(T+O atoms) and	d valence calculat	ion			
Si				0.001				
Be	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000
Sum T	1.000	1.000	1.000	1.001	1.000	1.000	1.000	1.000
Ti		0.001	0.010	0.003	0.002	0.001	0.002	0.002
Sn		0.001	0.002	0.004	0.000	0.000		
Al	1.967	1.948	1.917	1.903	1.967	1.958	1.976	1.978
Sc	0.001				0.001		0.001	
V			0.001	0.001	0.001			
Ga		0.001				0.002	0.001	0.001
Fe ³⁺	0.032	0.048	0.061	0.087	0.027	0.038	0.020	0.018
Fe ²⁺		0.001	0.009	0.002	0.002	0.001		0.001
Sum O	2.000	2.000	2.000	2.000	2.000	2.000	2.000	2.000
Al ₂ BeO ₄ (mol.%)	98.4	97.6	96.9	95.6	98.6	98.1	99.0	99.1
Fe ₂ ³⁺ BeO ₄ (mol.%)	1.6	2.4	3.1	4.4	1.4	1.9	1.0	0.9

bdl - below detection limit

Staněk 1981; Franz and Morteani, 1984; Černý et al., 1992). The metamorphic overprinting also caused partial dissolution–reprecipitation of accessory columbite–tantalite with extensive compositional and structural re-equilibration and homogenisation of primary magmatic Nb–Ta oxide minerals (Černý et al., 1992). Our observations corroborate metamorphic crystallisation of chrysoberyl by a breakdown of primary magmatic beryl, together with the coeval formation of metamorphic fibrolitic sillimanite, quartz and muscovite (Figs 3c, 4a). Detailed inspection of microtextures revealed the presence of small, commonly oval, relicts of

primary beryl in chrysoberyl or in quartz (Figs 3a,b,d, 5a) or growth of chrysoberyl crystals at the expense of primary magmatic beryl (Fig. 4c).

The following reactions have been suggested for the formation of new metamorphic assemblages from a primary magmatic beryl and associated minerals (Franz and Morteani, 1984, 2002):

$$beryl + 4albite + 4H^{+} = 3chrysoberyl + 18SiO_{2}/quartz \\ + 4Na^{+} + 2H_{2}O, \tag{1}$$

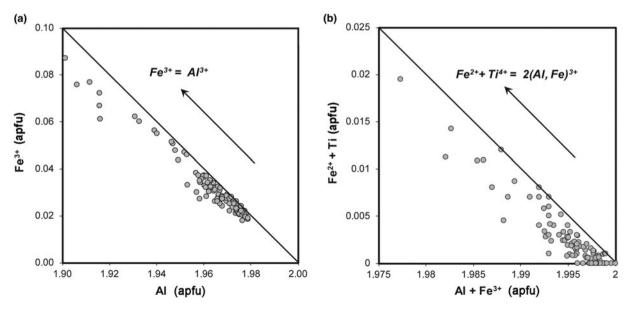


Figure 6. Substitution diagrams of chrysoberyl from the Maršíkov-Schinderhübel III pegmatite: (a) Fe³⁺ = Al³⁺; (b) Fe²⁺ + Ti⁴⁺ = 2(Al,Fe)³⁺.

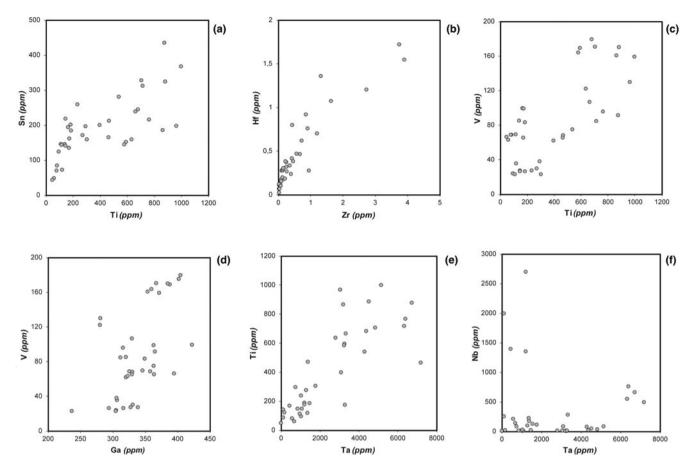


Figure 7. Correlation diagrams of trace elements from the Maršíkov–Schinderhübel III pegmatite (in ppm): (a) Sn vs. Ti; (b) Hf vs. Zr; (c) V vs. Ti; (d) V vs. Ga; (e) Ti vs. Ta; (f) Nb vs. Ta.

beryl
$$+4$$
albite $+2$ muscovite $+6H^+=3$ chrysoberyl $+3$ sillimanite $+21SiO_2/quartz$ $+4Na^++2K^++5H_2O$ and (2)

beryl + 9albite +
$$8H^++K^+=$$
 3chrysoberyl
+ muscovite + sillimanite + $29SiO_2$ /quartz
+ $9Na^++3H_2O$. (3)

Černý et al. (1992) have also proposed the following reactions:

beryl =
$$chrysoberyl + 6quartz + 2BeO$$
, (4)

2muscovite = 3sillimanite + 3quartz +
$$K_2O$$

+ $2H_2O$ and (5)

2albite = sillimanite
$$+ 5quartz + Na_2O$$
. (6)

The assemblage chrysoberyl+quartz \pm fibrolitic sillimanite \pm muscovite might be explained by the reactions 1 to 4. Aggregates of fibrolitic sillimanite (\pm tiny metamorphic muscovite) form

overgrowths on chrysoberyl crystals, or they occur in close vicinity (Figs 3c, 4a). Reaction 4 suggests that part of the Be from primary magmatic beryl escaped into fluids which might have facilitated precipitation of a distal chrysoberyl without close contact with primary magmatic beryl (Fig. 3e,f).

Experimental data and the presence of sillimanite clearly indicate high-temperature conditions for the beryl breakdown to a chrysoberyl and quartz assemblage, generally over 500-600°C (Franz and Morteani, 1984, 2002; Barton 1986; Barton and Young, 2002, and references therein). Černý et al. (1992) estimated temperatures of 570-630°C and pressures of 250-500 MPa for the metamorphic formation of chrysoberyl in the Maršíkov-Schinderhübel III pegmatite. Variscan prograde medium- to hightemperature metamorphism at $T \approx 540-660^{\circ}\text{C}$ and $P \approx 500-700$ MPa (Souček, 1978; René, 1983, Cháb et al., 1990; Schulmann and Gayer, 2000; Košuličová and Štípská, 2007; Schulmann et al., 2014) is probably responsible for the origin of metamorphic chrysoberyl and sillimanite in the Maršíkov-Schinderhübel III pegmatite. However, our textural and paragenetic observations do not support dividing the prograde metamorphic overprinting of the Maršíkov-Schinderhübel III pegmatite into two stages as Černý et al. (1992) proposed as an explanation for what appeared to be two generations of chrysoberyl. Chrysoberyl has also been found at several granitic pegmatites in the northern region around Česká Ves where it is commonly associated with rare sillimanite (e.g. Novák and Rejl, 1993). Hence, the metamorphic assemblage chrysoberyl + sillimanite in granitic pegmatites shows a regional

Table 2. Representative compositions (wt.%) and mineral formulae (apfu) of beryl from the Maršíkov pegmatite.

	SCH-7-	SCH-1E-	SCH-8-	SCH-4-	SCH-4-	SCH-4-
Sample #	14	10	6	8	9	10
Туре	Primary Fe-poor beryl			Secon	dary Fe-ric	h beryl
SiO ₂	65.27	65.62	65.63	63.24	62.91	63.39
TiO ₂	bdl	bdl	bdl	0.04	0.04	0.05
Al_2O_3	18.26	18.23	18.23	14.63	14.69	14.85
V_2O_3	0.09	bdl	bdl	bdl	bdl	bdl
Fe ₂ O ₃	0.09	0.67	1.19	1.14	0.68	0.83
FeO	0.46	0.21	bdl	3.06	3.41	3.17
MnO	bdl	0.07	bdl	0.22	0.17	0.20
ZnO	0.04	bdl	0.08	0.07	0.04	0.03
MgO	bdl	bdl	bdl	0.04	0.03	0.03
CaO	0.10	bdl	bdl	bdl	bdl	bdl
SrO	0.09	0.04	bdl	0.04	bdl	bdl
BeO (calc.)	13.63	13.71	13.75	13.15	13.09	13.18
Na ₂ O	0.16	0.23	0.15	1.40	1.55	1.44
Cs ₂ O	0.06	bdl	0.04	bdl	bdl	bdl
Total	98.25	98.78	99.07	97.03	96.61	97.17
Mineral form		on 18 O and	3 Be atom	ns, 8(<i>T1</i> + <i>O</i>) cations a	nd valence
calculation						
Si	5.978	5.976	5.965	6.006	6.003	6.005
Al <i>T1</i>	0.022	0.024	0.035	0.000	0.000	0.000
Sum T1	6.000	6.000	6.000	6.006	6.003	6.005
Ti				0.003	0.003	0.004
Al O	1.949	1.933	1.918	1.638	1.652	1.658
V	0.007					
Fe ³⁺	0.006	0.046	0.081	0.082	0.049	0.059
Fe ²⁺	0.035	0.016		0.243	0.272	0.251
Mn	0.000	0.005		0.018	0.014	0.016
Zn	0.003	0.000	0.005	0.005	0.003	0.002
Mg				0.006	0.004	0.004
Sum O	2.000	2.000	2.004	1.995	1.997	1.994
Be <i>T2</i>	3.000	3.000	3.000	3.000	3.000	3.000
Ca	0.010					
Sr	0.005	0.002		0.002		
Na	0.028	0.041	0.026	0.258	0.287	0.265
Cs	0.002		0.002			
Sum C	0.045	0.043	0.028	0.260	0.287	0.265
Al total	1.971	1.957	1.953	1.638	1.652	1.658
Sum	11.045	11.043	11.032	11.261	11.287	11.264
cations						

bdl – below detection limit. Other elements not listed: Cr, Ga, Sc, Y, K and Rb are also below the detection limit.

distribution generally consistent with a single Variscan prograde metamorphism in the examined region.

Investigation of mineral textures and composition also indicates a retrograde metamorphic stage in the Maršíkov–Schinderhübel III pegmatite. Chrysoberyl is replaced locally by secondary Fe-rich beryl, euclase, and late muscovite along veinlets and irregular rim domains (Figs 3a, 4b,e, 5). The textural relationships support partial replacement of euclase by bertrandite, whereas adjacent chrysoberyl is intact (Fig. 4b). For this retrograde stage, the following reactions are proposed:

10chrysoberyl + 28quartz +
$$2K_2O + 6H_2O$$

= 4euclase + 2beryl + 4muscovite, (7)

4euclase = bertrandite
$$+ 2Al_2O_3 + 2SiO_2 + H_2O$$
. (8)

Experimental studies indicate a lower stability of beryl at ~300–400°C, whereas euclase is stable at ~300–450°C, and bertrandite below 250°C at a pressure up to 300 MPa (Franz and

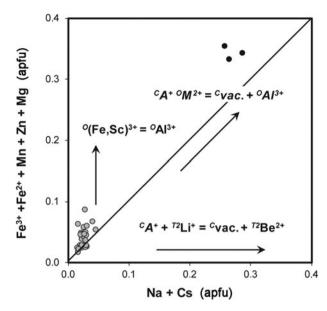


Figure 8. Substitution $Fe^{3+} + Fe^{2+} + Mn + Zn + Mg$ vs. Na + Cs diagram of beryl from the Maršíkov–Schinderhübel III pegmatite. Notes: primary magmatic beryl (grey circles), secondary Fe-rich beryl (black circles).

Table 3. Representative (wt.%) and mineral formulae (apfu) of euclase from Maršíkov pegmatite.

Sample #	SCH-1b-6	SCH-1b-7	SCH-1e-3	SCH-1e-9	SCH-2-12
SiO ₂	41.18	40.98	40.42	39.88	41.51
Al_2O_3	34.63	34.40	34.32	34.20	34.17
V_2O_3	bdl	bdl	bdl	0.05	bdl
Ga_2O_3	0.04	bdl	bdl	bdl	bdl
Fe ₂ O ₃	0.08	0.06	0.10	0.09	0.43
MnO	bdl	0.03	0.07	0.03	bdl
ZnO	0.09	0.03	0.07	0.11	bdl
BeO (calc.)	17.16	17.01	16.91	16.75	17.14
MgO	bdl	bdl	bdl	bdl	0.02
CaO	0.02	0.02	0.07	0.05	0.10
Na ₂ O	0.09	bdl	bdl	bdl	bdl
K ₂ O	0.02	bdl	bdl	bdl	0.07
H ₂ O (calc.)	6.18	6.11	6.09	6.00	6.17
F	bdl	0.03	bdl	0.08	bdl
Total	99.51	98.67	98.05	97.23	99.61
Mineral form	nulae based o	n 5 anions, 1(OH+F) and 1	Be atom	
Si	0.999	1.003	0.995	0.991	1.008
Al	0.001	0.000	0.005	0.009	0.000
Sum T	1.000	1.003	1.000	1.000	1.008
Al	0.989	0.992	0.991	0.992	0.978
V				0.001	
Ga	0.001				
Fe	0.002	0.001	0.002	0.002	0.008
Mn		0.001	0.001	0.001	
Mg					0.001
Zn	0.002	0.001	0.001	0.002	
Sum O	0.993	0.995	0.996	0.998	0.987
Be	1.000	1.000	1.000	1.000	1.000
Ca	0.001	0.001	0.002	0.001	0.003
Na	0.004				
K	0.001				0.002
Sum A	0.005	0.001	0.002	0.001	0.005
ОН	1.000	0.998	1.000	0.994	1.000
F		0.002		0.006	
Sum X	1.000	1.000	1.000	1.000	1.000

bdl – below detection limit. Other elements not listed: Ti, Sn, Cr, Sc, Y, Ni, Mg, Sr, Rb, Cs and Cl are also below the detection limit.

Table 4. Representative (wt.%) and mineral formulae (apfu) of bertrandite from the Maršíkov pegmatite.

Sample	SCH-1-11	SCH-1e-4	SCH-3-18	SCH-3-19
SiO ₂	48.34	49.60	49.56	49.69
TiO ₂	bdl	0.03	0.04	bdl
Al_2O_3	1.56	0.14	0.28	0.09
Ga_2O_3	0.09	0.02	0.05	bdl
Cr_2O_3	0.06	bdl	0.02	bdl
V_2O_3	bdl	0.02	bdl	bdl
Sc_2O_3	bdl	0.02	bdl	bdl
Fe ₂ O ₃	0.03	0.06	0.07	bdl
ZnO	bdl	0.04	0.05	0.08
BeO (calc.)	41.92	41.56	41.69	41.54
CaO	0.06	bdl	bdl	bdl
SrO	0.03	0.04	bdl	bdl
Na ₂ O	0.13	0.01	0.01	0.02
K ₂ O	0.07	0.01	0.08	0.04
H ₂ O (calc.)	7.55	7.47	7.51	7.47
F	bdl	0.02	bdl	0.01
Total	99.83	99.05	99.35	98.94
Mineral formu	lae based on 9 a	nions, 2B cations	s, 4 Be atoms and	d 2(OH+F)
Si	1.920	1.988	1.980	1.992
Ti		0.001	0.001	
Al	0.073	0.007	0.013	0.004
Ga	0.002	0.001	0.001	
Cr	0.002		0.001	
V		0.001		
Sc		0.001		
Fe	0.001	0.002	0.002	
Zn		0.001	0.002	0.002
Sum B	2.000	2.000	2.000	2.000
Be	4.000	4.000	4.000	4.000
Ca	0.002			
Sr	0.001	0.001		
Na	0.010	0.001	0.001	0.001
K	0.004	0.001	0.004	0.002
Sum A	0.017	0.003	0.005	0.003
OH	2.000	1.997	2.000	2.000
F		0.003		0.000
Sum X	2.000	2.000	2.000	2.000

bdl – below detection limit. Other elements not listed: Sn, Mn, Ni, Mg, Rb, Cs and Cl are also below the detection limit.

Table 5. The unit-cell parameters of chrysoberyl from the Maršíkov pegmatite calculated from powder XRD and compared with published data.

Locality/Reference	a (Å)	b (Å)	c (Å)	V (Å ³)
Maršíkov (this study)	4.4289(8)	9.413(2)	5.480(1)	228.457
Maršíkov (Dostál, 1969)	4.430(2)	9.414(8)	5.477(4)	228.413
Hazen and Finger (1987)	4.424(1)	9.396(1)	5.471(2)	227.5(1)

Morteani 1981, 2002; Barton 1986; Barton and Young, 2002; Grew, 2002, and reference therein). However, a recent study of secondary Be minerals and their mineral assemblages from granitic pegmatites (Novák *et al.*, 2023) suggests the stability of bertrandite up to $T \approx 300^{\circ}$ C. Consequently, this late retrograde metamorphic event occurred at $T \approx 500$ to 200°C and $P \leq 250$ MPa. The existence of the Permian post-Variscan thermal overprinting (~280 to 260 Ma) is documented in the Hrubý Jeseník Mountains by K–Ar mica and U–Th–Pb monazite radiometric age determinations; the event was probably associated with renewed fluid activity along the Sudetic fault system (Schulmann *et al.*, 2014).

Conclusions

A study of the compositional variation of chrysoberyl indicates the following principal substitution mechanisms in the octahedral sites: (1) $Fe^{3+} = AI^{3+}$ and (2) $Fe^{2+} + Ti^{4+} = 2(AI,Fe)^{3+}$. Moreover, the characteristic trace elements incorporated in the chrysoberyl structure are Ga, Sn and V, whereas unusually high Ta and Nb concentrations can be attributed to nano- to micro-inclusions of Nb–Ta oxide minerals (especially columbite–tantalite).

The Maršíkov–Schinderhübel III granitic pegmatite represents a classic example of a beryl–columbite granitic pegmatite affected by extensive metamorphic overprinting. The first prograde stage of this metamorphism produced chrysoberyl, quartz, sillimanite muscovite at the expense of primary magmatic beryl, albite and muscovite at amphibolite-facies conditions (~600°C and 250–

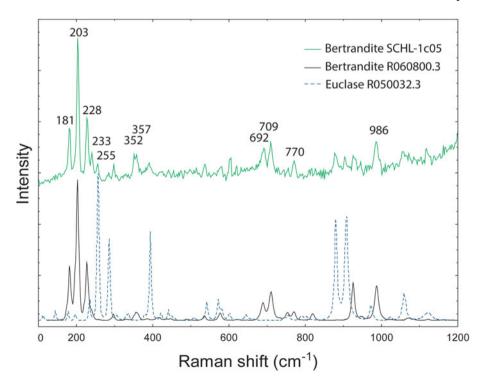


Figure 9. Micro-Raman spectrum of bertrandite with admixture of euclase from the Maršíkov–Schinderhübel III pegmatite.

500 MPa). The subsequent second retrograde stage, probably related to fluid activity along the fault system, resulted in partial alteration of chrysoberyl and crystallisation of secondary Fe-rich beryl, euclase, bertrandite and late muscovite at low-grade conditions (\sim 200–500°C and \leq 250 MPa).

Compositions, mineral assemblages and textural relationships of the Be minerals from the Maršíkov–Schinderhübel III granitic pegmatite support the concept that they might serve as useful geochemical and petrological mineral indicators of the conditions of their formation (Barton and Young, 2002; Černý, 2002; Franz and Morteani, 2002; Grew, 2002).

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Competing interests. The authors declare none.

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