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Depressurization and boiling of a single magmatic fluid as a mechanism for tin-tungsten deposit formation

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ABSTRACT

Tin (Sn) and tungsten (W) mineralization are often associated with each other in relation to highly evolved granites, but economical ore grades are restricted to rare global occurrences and mineralization styles are highly variable, indicating different mechanisms for ore formation. The Sn-W Zinnwald deposit in the Erzgebirge (Germany/Czech Republic) in the roof zone of a Variscan Li-F granite hosts two contrasting styles of mineralization: 1) cassiterite (Sn) in greisen bodies and 2) cassiterite and wolframite (W) in predominantly sub-horizontal quartz-rich veins. The relative timing and causes for ore formation remain elusive. Studies of fluid inclusion assemblages in wolframite, cassiterite and quartz samples from greisen and veins by conventional and infrared microthermometry and LA-ICP-MS analyses have revealed compelling evidence that all elements required for the formation of the Zinnwald Sn-W deposit were contained in a single parental magmatic-hydrothermal fluid that underwent two main processes: 1) fluid-rock interaction during Sn-greisen formation and 2) depressurization and vapor loss leading to ore precipitation in quartz-Sn-W veins. The results also show that fluid inclusion assemblages in ore minerals can document fluid processes that are absent in the fluid inclusion record of gangue minerals. The study further highlights the role of phase separation in the formation of W-rich Sn-deposits and indicates that W-deposits in distal parts of evolved granites may be restricted to fluids derived from deeper-seated plutons.

INTRODUCTION

Natural enrichments of tin (Sn) and tungsten (W) to economic grades are typically associated with evolved A- or S-type granites in major orogenic settings (e.g., Romer and Kroner (2016) and references therein). Sn, W or Sn-W mineralization are hosted in metasomatized roof zones (greisen) and/or hydrothermal veins, which can be

located proximal within the intrusion, e.g., Erzgebirge, Germany/Czech Republic (Stemprok, 1967) and East Kemptville, Canada (Halter et al., 1998), or may extend to more distal parts into the surrounding rocks, e.g., Panasqueira, Portugal (Kelly and Rye, 1979) and Pasto Bueno, Peru (Landis and Rye, 1974). However, fluid sources and pathways as well as their temporal and spatial evolution which lead to Sn-W ore precipitation often remain debated.

Within the same deposit, precipitation of cassiterite, wolframite and other ore minerals have occurred contemporaneously or successively, indicating that several geologic controls can trigger ore deposition. Greisen formation has mainly been attributed to hydrolysis of feldspars (Stemprok, 1987; Stemprok and Sulcek, 1969) by magmatic fluids, with pH decrease leading to Sn precipitation (Schmidt, 2016). Sn-vein formation at Mole Granite, Australia (Audétat et al., 2000) and W-Sn-vein formation at Vaulry, France (Vallance et al., 2001) are related to mixing of magmatic and meteoric fluids. W-mineralization at Panasqueira has been attributed to fluid-rock reactions of W-rich, Fe-poor magmatic fluids with Fe-rich metasedimentary host rocks (Lecumberri-Sanchez et al., 2017). Phase separation has further been identified as a potential mechanism for ore deposition, albeit with a subordinate efficiency to form an economic ore deposit (Heinrich, 1990). About half of the global W production comes from vein, stockwork, breccia, and porphyry deposits and only 5% from disseminated greisen mineralization, which in contrast is the main source for Sn (Pitfield et al., 2011).

Infrared microthermometry and LA-ICP-MS analyses of fluid inclusion assemblages (FIA) in ore minerals such as cassiterite and wolframite can provide direct insights to temperature, salinity and chemistry of ore-forming fluids (Lüders, 2017), but LA-ICP-MS analyses of ore-hosted fluid inclusions (FIs) require a very precise documentation of the location of fluid inclusions prior to ablation (Kouzmanov et al., 2010).

THE ZINNWALD SN-W DEPOSIT

The Zinnwald/Cínovec deposit is located on both sides of the German/Czech border in the eastern part of the Erzgebirge, which is part of the Saxothuringian Zone of the Variscan Orogeny (Fig. 1). The deposit is related to the evolved Li-F Zinnwald granite (Förster et al., 1998) and hosts two mineralization styles: 1) Sn in greisen (Fig. 1B) and 2) proximal Sn-W-veins (Fig. 1D) within the metasomatized roof zone of the granitic intrusion (Appendix for detailed geology). Fluid inclusion microthermometry in quartz and cassiterite from tin-tungsten deposits in the Erzgebirge showed a predominance of intermediate-density fluids and temperatures between 360 and 400 °C for ore formation (Thomas, 1982). Studies of wolframite-hosted fluid inclusions from the Zinnwald deposit yielded homogenization temperatures between 300 and 360 °C and variable salinity (Lüders, 1996). Few LA-ICP-MS measurements of fluid inclusions in quartz from the Zinnwald Sn-W deposit have been performed in rare boiling assemblages at higher temperatures of ca. 470 °C (Heinrich et al., 1999). However, the fluid evolution in Zinnwald and ore formation remains debated.

For this study, we collected samples from quartz veins from the German part of the mine at the northern flank of the granite (Fig. 1A). Additional historic samples

containing euhedral crystals of cassiterite, wolframite and quartz were provided from mineral collections. Fluid inclusion in 25 selected samples of quartz and ore minerals were studied by microthermometry (convectional and infrared), isotope ratio mass spectrometry and LA-ICP-MS (see Appendix for detailed method description).

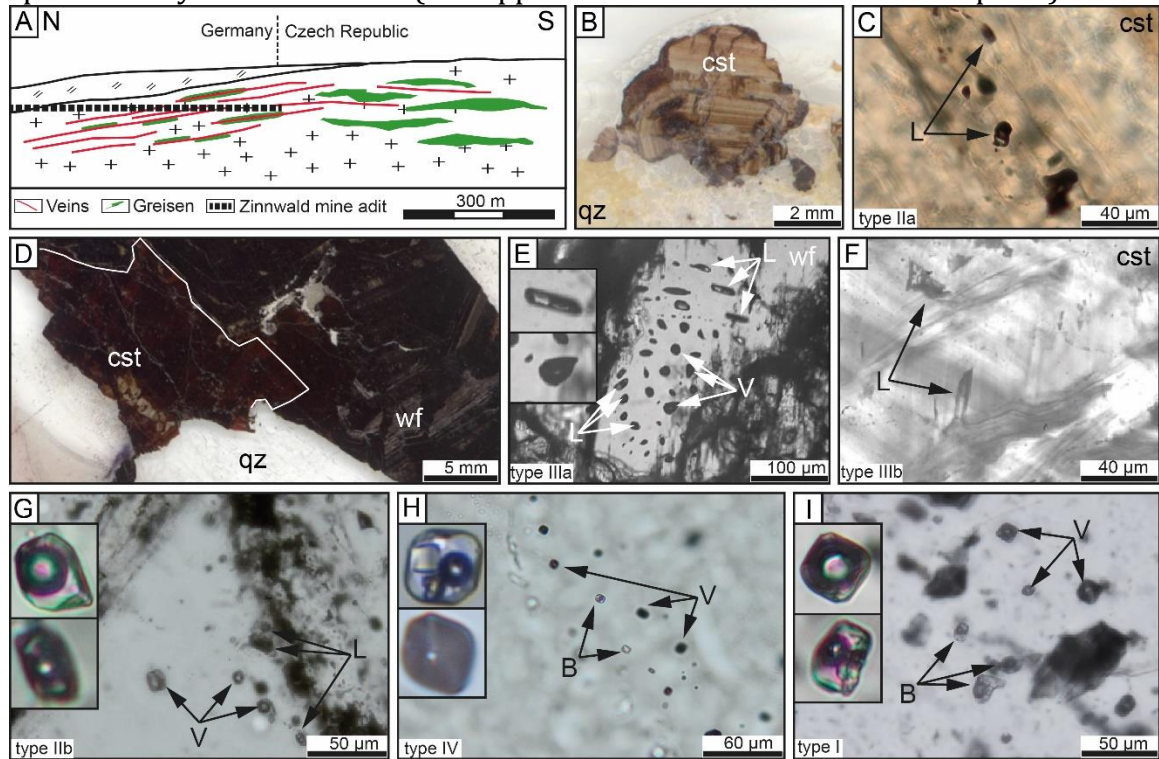


Figure 1. A: Schematic cross section through the roof zone of the Zinnwald granite showing the sampling area (adit) with greisen and vein-type mineralization. B + C: Greisen sample with quartz (qz) and cassiterite (cst) showing growth zones with primary liquid-rich (L) inclusions. D: Wolframite (wf) intimately intergrown with cassiterite indicating contemporaneous deposition in the vein. E: Wolframite with a boiling assemblage of liquid-rich and vapor-rich (V) inclusions. F: Cassiterite with intermediate density inclusions orientated along growth zones. G: Fluid inclusion assemblage in the wake of a quartz growth zone showing different homogenization behavior: L to liquid, V to vapor. H: Trail of a boiling assemblage with pseudosecondary brine-type inclusions (B) in quartz. I: High-temperature boiling assemblage in a quartz growth zone with hypersaline brines.

RESULTS

The studied quartz samples are often recrystallized and host abundant secondary fluid inclusions. Rare primary and pseudosecondary fluid inclusions and fluid inclusion assemblages hosted in quartz, cassiterite and wolframite were classified after Roedder (1984) and define four different inclusion types (Fig. 2A). Type I high-temperature brine inclusions from boiling assemblages hosted in a re-analyzed quartz sample from Heinrich et al. (1999) and in one quartz sample from the BGR collection yield salinities of ca. 40 wt.% NaCl equiv. and homogenization temperatures between 420 and 490 °C (Fig. 1I). The most common primary fluid inclusions (type II) decorate growth zones and are aqueous two-phase inclusions with liquid to vapor ratios of ~1:1. They are hosted in greisen (quartz and cassiterite: type IIa; Fig. 1C) as well as in vein quartz (type IIb; Fig. 1G). Type II inclusions commonly homogenize into the liquid phase between 350 °C and 410 °C, but in rare

FIAs they homogenize either into the liquid or vapor phase (Fig. 1G) or even show critical homogenization. Salinities of type II inclusions range between 1 and 8.5 wt.% NaCl equiv. Type III inclusions are aqueous two-phase inclusions hosted in wolframite (type IIIa; Fig. 1E) and cassiterite (type IIIb; Fig. 1F) in veins. Most type IIIa inclusions are liquid-rich, have homogenization temperatures between 340 and 350 °C and salinities of 10 – 12 wt.% NaCl equiv. Type IIIa inclusions are locally found in boiling assemblages (Fig. 1E). Type IIIb inclusions in cassiterite have lower homogenization temperatures of ca. 335 °C and higher salinities of ~14 wt.% NaCl equiv. even though cassiterite is intimately intergrown with wolframite. Type III inclusions are not contained in quartz, not even in quartz intergrown with wolframite (Fig. 1D). Rare type IV fluid inclusion low-temperature boiling assemblages were observed in two samples from a quartz vein (Fig. 1H). The samples host abundant small vapor-rich inclusions besides brine inclusions which mostly contain two to three solid phases and have salinities around 30 wt.% NaCl equiv. and homogenization temperatures of ca. 300 °C.

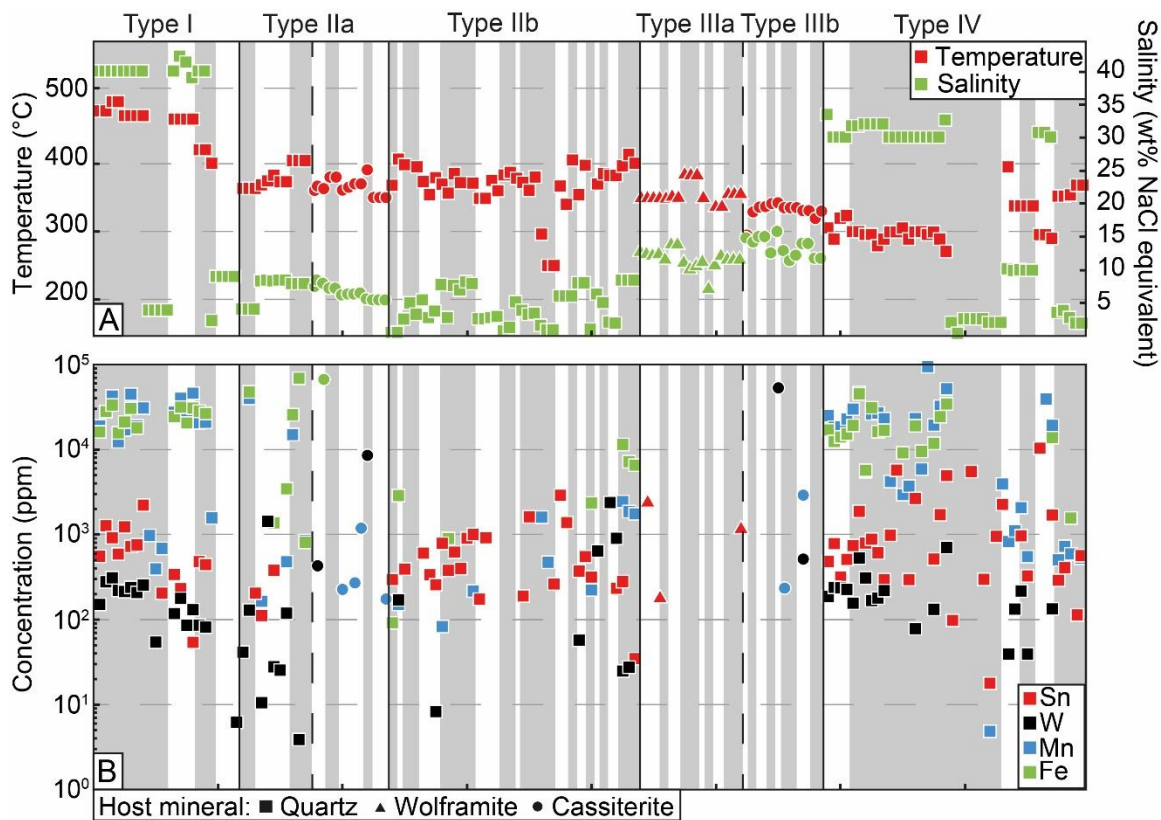


Figure 2. A: Homogenization temperatures and salinities of different fluid inclusion types (I – IV) in quartz (squares), cassiterite (circles) and wolframite (triangles). Grey and white fields group individual fluid inclusion assemblages. B: Element contents in fluid inclusions by LA-ICP-MS.

LA-ICP-MS analyses show that all elements necessary for the formation of cassiterite (Sn) and wolframite (Mn, Fe, W) can be detected in all inclusion types (Fig. 2B). Sn contents are slightly higher in the boiling assemblages (I, IV) than in type IIb

inclusions. Wolframite has higher traces of Sn, but some type IIIa inclusions have detectable Sn contents of up to 2400 ppm. Relevant metals for the formation of wolframite are more enriched in brine-type inclusions and have similar contents in greisen (IIa) and vein quartz (IIb).

The different fluid inclusion types show a strong similarity in chemical compositions, suggesting a common source fluid, with variations mainly depending on different salinities due to phase separation and fractionation. A single fluid source is best documented by various almost linear positive correlations, e.g., Cs/Na versus Rb/Na ratios and As versus Cs contents (Fig. 3A + B) and homogeneous $\delta^{13}\text{C}_{\text{CO}_2}$ values of fluid inclusions in quartz and cassiterite between $\delta 5.5$ and $\delta 7$ ‰, which is consistent with a magmatic fluid source (e.g. Fuex and Baker, 1973). The geochemical data further show that fluid inclusions in all type II inclusions have similar element contents whereas the FI from boiling assemblages (type I, III, IV) form a distinct cluster of high element concentrations (see Appendix for statistical values).

FLUID EVOLUTION

Type IV FIA allow a direct estimate of the fluid pressures during vein formation because they are tied to the two-phase liquid-vapor (LV) coexistence curve, yielding fluid pressures of $\sim 60 - 100$ bar (Fig. 3C). Type IIb inclusions with variable salinities lack boiling assemblages and yield minimum entrapment pressures between 150 and 330 bar. Their variable homogenization behavior indicate that they were also likely trapped at or near the LV-coexistence and that the salinity variations of 1 – 10 wt.% NaCl equiv. reflect gentle boiling. Since all sampled veins represent a similar depth level (Fig. 1A) and type II and IV inclusions occur in the same vein, the pressure variation with a factor 1.5 – 5.5 can best be explained by transition from a (nearly) lithostatic to a (nearly) hydrostatic pressure regime, suggesting a paleodepth of $\sim 1 - 2$ km in agreement with estimates for the granite intrusion depth (Dolejš and Stempok, 2001). Lithostatic fluid pressure conditions have been inferred as a possible mechanism for the formation of sub-horizontal veins by hydraulic fracturing during fluid exsolution from a crystallizing magma chamber (Breiter et al., 2017; Foxford et al., 2000).

Type I inclusions hosted in historic quartz samples yield higher pressure conditions between 250 and 450 bar during fluid entrapment, suggesting sample origin from deeper parts of the deposit. The high temperatures partially narrow the temperature gap between an assumed solidus temperature of the granite of $\sim 650^\circ\text{C}$ (Johannes, 1984) and the average mineralization temperature of 380°C .

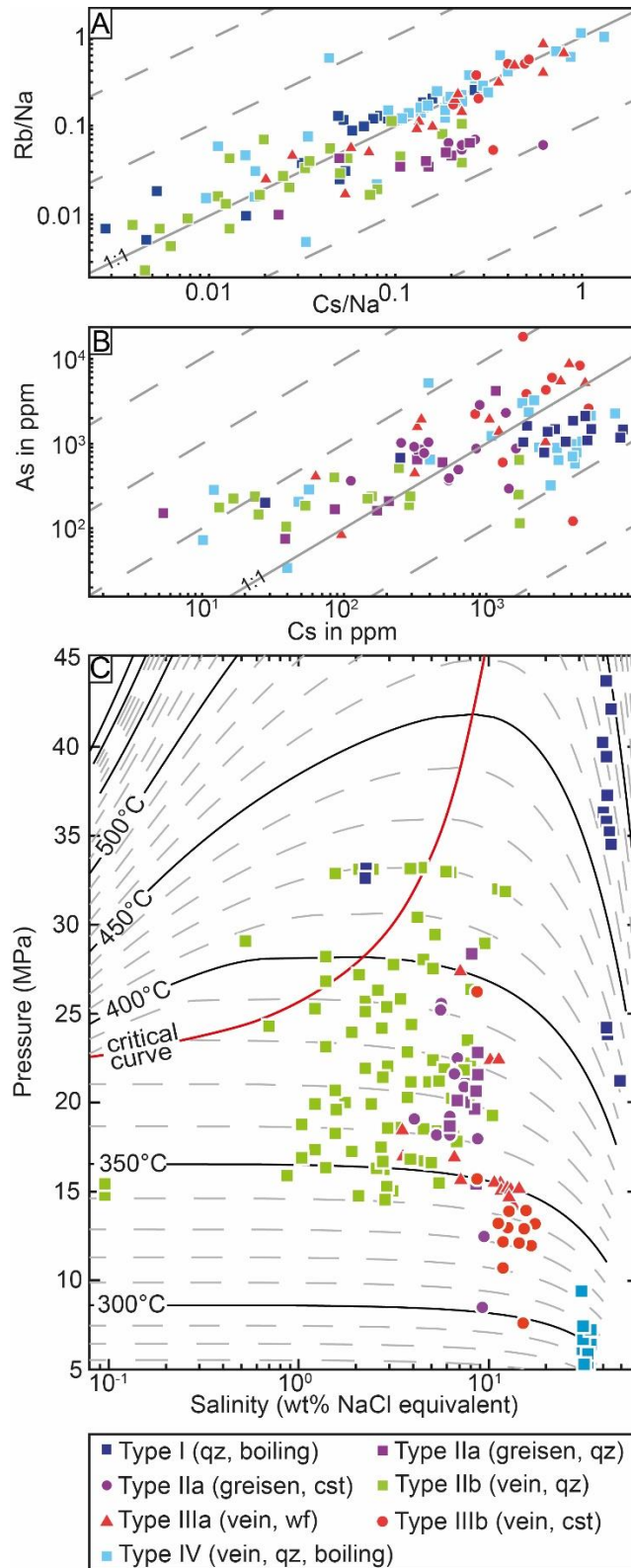


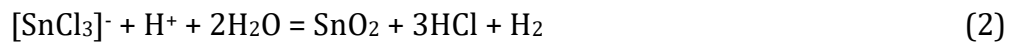
Figure 3. A + B: Cs/Na versus Rb/Na ratios and Cs versus As contents of different fluid inclusion types showing near-linear trends. **C:** Pressure estimates from homogenization temperatures and salinities. Fluid inclusions of type IIa in Greisen overlap with type IIb inclusions. Fluid inclusions of type IIIa and IIIb in vein wolframite and cassiterite form a distinct field between type II and type IV brine inclusions. Fluid pressures, phase relations and isotherms are calculated after Driesner and Heinrich (2007).

ORE PRECIPITATION

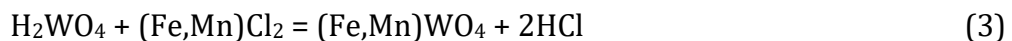
The type III inclusions in ore minerals from vein mineralization follow the same trend of decreasing pressures and temperatures and increasing salinities along the two-phase coexistence field. Their entrapment conditions are positioned in the middle between the inclusions representing the lithostatic (type II) and the hydrostatic (type IV) endmembers of the fluid evolution in the veins. This interpretation is confirmed by the presence of boiling FIAs in wolframite (type IIIa) and their geochemical similarities to other boiling FIAs. The FIA in the intergrown wolframite-cassiterite sample form slightly different types (IIIa and IIIb, respectively), which suggests a rapid, dynamic process during ore deposition, such as depressurization during fracture opening. A transient pressure drop from nearly-lithostatic to sub-hydrostatic conditions leads to partial loss of the high-enthalpy vapor phase, resulting in cooling and salinity increase of the remaining brine.

The similarities in chemical composition and partial overlap of fluid inclusions types IIa (greisen) and IIb (vein quartz) suggest a simultaneous formation of vein and greisen in Zinnwald, which is in line with geological observations at the mine where quartz veins can be hosted by greisen, granite (not greisenized) or partially by granite and greisen.

Our interpretation suggests two contrasting processes of Sn-W deposition for greisen and vein mineralization: (1) fluid-rock reaction and (2) phase separation. Sn can be transported as Sn(II) or Sn(IV) by complexation with Cl⁻ in the fluid whereas W is in solution as WO₄²⁻. Therefore, SnO₂ (cassiterite) can be precipitated by removing HCl from the fluid system (Schmidt, 2016):



Wolframite precipitation needs additional cations (here, Fe and Mn) which are also mostly transported as complexes with Cl⁻ (Heinrich, 1990):



Greisenization (hydrolysis of feldspars and biotite) moves the Sn-reactions (1, 2) to the right and can therefore precipitate cassiterite, whereas W can still stay in solution, e.g., if Fe and Mn are consumed for the formation of zinnwaldite (Tischendorf et al., 1997). During fluid phase separation volatile elements (e.g., HCl, Cl⁻, H⁺) preferentially partition into the vapor phase (Bischoff et al., 1996; Simonson and Palmer, 1993) which can readily leave the system due to its low density, leading to a change in the chemical composition of the fluid and therefore to cassiterite and simultaneous wolframite precipitation (Heinrich, 1990).

Phase separation during vein opening may be episodic, but the presence of a brine phase and the absence of pure vapor inclusions exclude extreme pressure drops during flashing events (Clark and Williams-Jones, 1990; Moncada et al., 2017). Ore

precipitation due to phase separation with the proposed mechanism will be most efficient at relatively low pressures where the vapor phase has a very low density. This relation suggests that greisen deposits with Sn-W-rich veins are more favorably associated with shallow intrusions, while fluids exsolved from deeper intrusions may be capable of avoiding wolframite precipitation by phase separation and allow metal transport to more distal parts, where fluids can interact with the surrounding rocks (e.g. Polya, 1989).

CONCLUSIONS

Our fluid inclusion study suggests that contrasting mineralization styles at the Sn-W Zinnwald deposit, Erzgebirge formed from a single magmatic fluid:

1. Feldspar hydrolysis during fluid-rock interaction in the roof of an evolved granitic intrusion leads to the formation of cassiterite-bearing greisen bodies.
2. Transient changes of the pressure regime from lithostatic to (sub-)hydrostatic conditions due to vein opening leads to phase separation and vapor loss, triggering coeval precipitation of wolframite and cassiterite in quartz veins within the greisenized cupola.

The fluid inclusion data, together with geological observations at the mine, suggest that both precipitation mechanisms happen roughly at the same time. The results of this study also show that fluid inclusion assemblages in ore minerals can form distinct compositional fields, which may be overlooked or not even documented in primary growth zones in coexisting vein quartz, highlighting the importance of fluid inclusion analyses in ore minerals as direct evidence for ore formation in dynamic hydrothermal systems.

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1GSA Data Repository item 2017xxx, Appendix DR1 (regional geology, methods, samples and further data) and Appendix DR2 (fluid inclusion data), is available online at <http://www.geosociety.org/datarepository/2017/> or on request from editing@geosociety.org.