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Cassiterite U-Pb geochronology constrains magmatic-hydrothermal evolution in complex evolved granite systems: The classic Erzgebirge tin province (Saxony and Bohemia)

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ABSTRACT

Laser ablation–inductively coupled plasma–quadrupole mass spectrometry analysis on hydrothermal cassiterite was applied to solve the long-standing debate on the age of the central European Erzgebirge-Krušné hory tin province of eastern Germany and the western Czech Republic. Cassiterite samples from the Sadisdorf, Ehrenfriedersdorf, Altenberg, Cinovec-Zinnwald, and Krupka tin deposits have U-Pb ages that overlap within the 2σ uncertainty range, and define a relatively narrow time window of 326 ± 3 to 320 ± 3 Ma. These dates also define the minimum age of the associated highly evolved alkali feldspar granite intrusions, which so far could not be reliably dated by conventional U-Pb, Sm-Nd, Rb-Sr, or ^{40}Ar-^{39}Ar techniques due to high-U metamict zircons and resetting by later thermal events. The time span of ca. 326–320 Ma characterizes the age range of regional rare-metal granite magmatism and associated hydrothermal tin mineralization in the Erzgebirge as a result of extensional collapse of the Variscan orogen.

INTRODUCTION

Major hydrothermal tin deposits are spatially and chemically related to highly evolved granitic rocks (Taylor, 1979; Lehmann, 1990). These felsic rocks commonly underwent pervasive late magmatic and subsolidus alteration. The conventional radioisotopic dating methods of hydrothermal tin deposits and associated granitic rocks include zircon U-Pb, whole-rock (WR)/mineral Rb-Sr, Sm-Nd, K/^{40}Ar-^{39}Ar, and molybdenite Re-Os isotope analysis. Zircon in highly fractionated granitic rocks is often radiation-damaged by recoil of α particles from elevated U content, which results in a disturbed U-Pb system (Davis and Krogh, 2001; Romer, 2003). Rb-Sr, Sm-Nd, and K/^{40}Ar-^{39}Ar systems have relatively low closure temperatures, and are easily overprinted and reset by late-stage hydrothermal alteration and later thermal events (Romer et al., 2007). Molybdenite from tin deposits may have low Re but high common Os contents, and the Re-Os system can occasionally be reset by Re loss during dissolution and/or precipitation reactions of molybdenite (McCandless et al., 1993). Cassiterite is the common ore mineral of tin deposits and has U only in the low parts per million range. Cassiterite U-Pb dating can directly constrain the tin mineralization event (Gulson and Jones, 1992; Yuan et al., 2008). The U-Pb isotopic closure temperature of cassiterite is inferred to be higher than the granite solidus temperature and its usual crystallization temperature of 300–450 °C (Zhang et al., 2011). Therefore, the U-Pb isotope system of cassiterite is particularly suitable for dating rare-metal granites and their associated hydrothermal tin deposits (Li et al., 2016). In addition, cassiterite also occurs in the heavy-mineral spectrum of clastic sediments, and its age information can then be used to trace sedimentary sources.

The Erzgebirge or Krušné hory (ore mountains in German or Czech) in the northwestern part of the Bohemian Massif of eastern Germany and the western Czech Republic is the birthplace of modern mining geology (Agricola, 1546), and its variety of hydrothermal ore deposits provided the basis for ground-breaking concepts in economic geology (Werner, 1791; von Cotta, 1859; Sandberger, 1885). Silver mining of the polymetallic veins around Freiberg started in A.D. 1168, and tin mining from placers at Krupka and Zinnwald dates to the Bronze Age. After World War II, the Erzgebirge-Krušné hory became the site of major uranium mining. The great scientific tradition of the Erzgebirge and extensive metal exploration and modern research over the past 50 yr make this region a particularly well studied granite-related metal province. However, in spite of the long-standing research history of the Erzgebirge, there are unresolved fundamental problems, such as the exact timing of tin and rare metal mineralization and that of granite magmatism (Förster and Romer, 2010). The classical and prevailing view on the granite magmatism and its associated magmatic-hydrothermal ore deposits is a grouping in older (330–320 Ma) and younger granite suites (310–290 Ma), based on geochemical and mineralogical features and field geological relationships, separated by a poorly constrained hiatus of as much as 20 m.y. (Lange et al., 1972; Tischendorf, 1987; Gerstenberger, 1989; Breiter et al., 1999; Kempe et al., 2004). This concept has been challenged by some carefully prepared U-Pb isotope analyses on zircon and uraninite inclusions in mica (Romer et al., 2007, 2010) and Re-Os ages on molybdenite (Romer et al., 2007; Ackerman et al., 2017). The chronological problems are due to the highly evolved nature of the granitic rocks, which invariably have a hydrothermal overprint, i.e., disturbance of the WR/mineral Sm-Nd, Rb-Sr, K/^{40}Ar-^{39}Ar systems (Tichomirowa and Leonhardt, 2010), as well as the open U-Pb isotope system of metamict zircon (Kempe et al., 2004; Romer et al., 2010). We here present U-Pb isotope data on hydrothermal cassiterite from some classic tin deposits of the Erzgebirge that firmly constrain the timing of the tin mineralization and the underlying granite magmatism as one relatively brief period of ~6 m.y. (Romer et al., 2007) when both older and younger granites form within the analytical uncertainty range of their radioisotopic ages.

GEOLOGICAL BACKGROUND

Cornwall (UK), the Bohemian Massif, the Iberian Massif, and the French Massif Central are the four key tin provinces in the European Variscan orogen (Fig. 1). The Erzgebirge-Krušné hory tin province is the second largest after Cornwall. The basic geodynamic setting is in the

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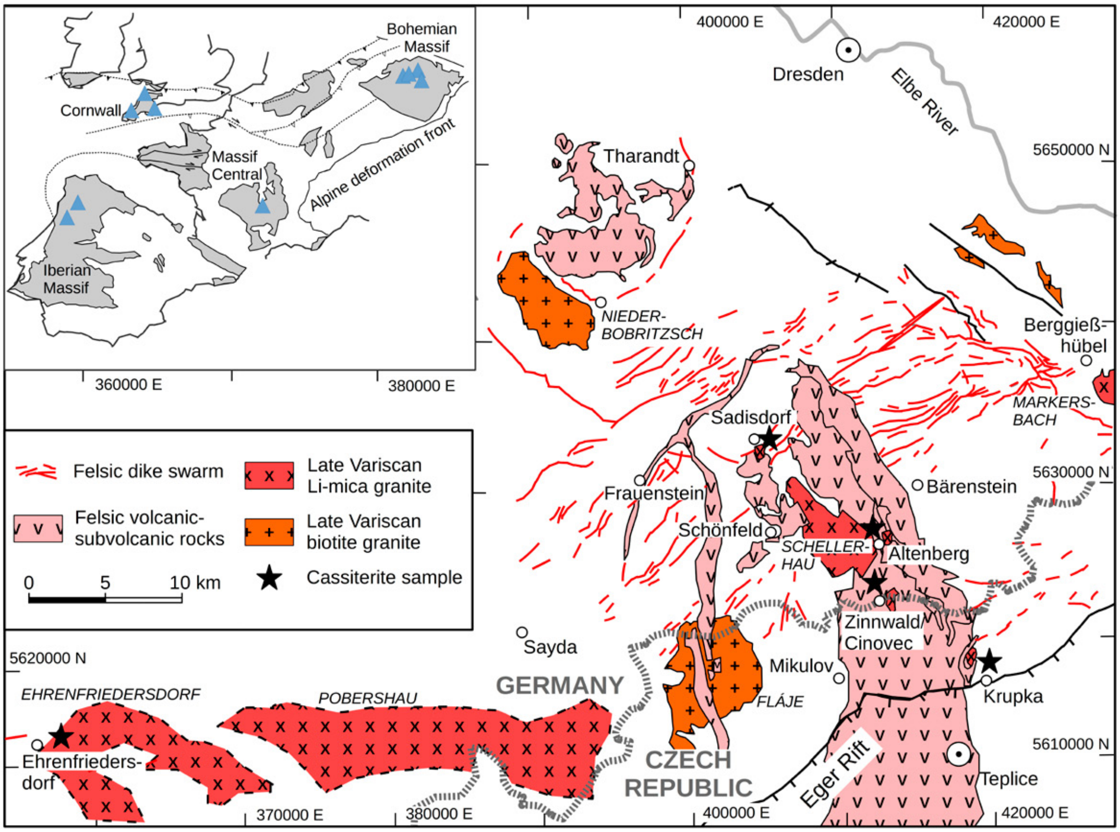

Figure 1. The eastern and central Erzgebirge-Krušné hory metal province in eastern Germany and western Czech Republic, with focus on late-collisional to post-collisional highly evolved Variscan granite magmatism, mapped as late Variscan biotite granite and late Variscan Li-mica granite. Note that the alkali feldspar granite intrusions in the central Erzgebirge are mainly unexposed (stippled outline); their shallow subsurface setting is confirmed by many drillholes from uranium and tin exploration in the former German Democratic Republic. Cassiterite sample locations are marked by asterisk. Inset map locates major tin deposits in the European Variscan orogenic belt (gray shading), marked by blue triangles.

Variscan orogen of western and central Europe, with the Erzgebirge at the leading edge of pre-Variscan Gondwana during its collision with Laurussia in the late Paleozoic (Matte, 1986). Based on geochemical and mineralogical composition, two major late-collisional or post-collisional granite units have been identified, i.e., low-F biotite granite and high-F Li-mica granite (Fig. 1). Both granite units are cut by felsic dikes, and the granites in the Altenberg area are part of a large volcanoplutonic caldera structure (Förster et al., 1999). Li-mica granite (alkali feldspar granite) is highly fractionated and genetically associated with the major tin deposits, e.g., Sadisdorf, Ehrenfriedersdorf, Altenberg, Cinovec-Zinnwald, and Krupka (Fig. 1). The available WR/mica Rb-Sr, mica K/^{40}Ar-^{39}Ar, and U-Pb chemical and isotope dilution–thermal ionization mass spectrometry (ID-TIMS) age data on zircon cover a large time span from 332 to 291 Ma for the high-F Li-mica granites (Kempe et al., 2004; Romer et al., 2007). Recent ^{40}Ar-^{39}Ar age data on Li-mica from mica-quartz-topaz greisen at Sadisdorf, Zinnwald-Cinovec, Ehrenfriedersdorf, and Krupka converge over a range of 315–310 Ma (Seifert et al., 2011; Seifert and Pavlova, 2016), which is, however, significantly younger than the available molybdenite Re-Os model ages of 322–315 Ma from the same deposits (Romer et al., 2007; Ackerman et al., 2017). Gerstenberger (1989) presented a Rb-Sr WR errorchron age of 305 ± 3 Ma (Sr_i = 0.6890 ± 165) for the Altenberg Li-mica granite, consistent with the ^{40}Ar-^{39}Ar plateau ages (309–307 Ma) of biotite (Seltmann and Schilka, 1995), but younger than the Re-Os model ages of 317.9 ± 2 Ma and 323.9 ± 2 Ma for individual molybdenite samples from massive greisen and quartz veinlets (Romer et al., 2007).

SAMPLING AND ANALYTICAL METHODS

We sampled cassiterite from five major tin deposits that can be regarded as representative of the Erzgebirge-Krušné hory province: (1) Sadisdorf Sn-W greisen-vein deposit, sample SD-1 from the open pit; (2) Ehrenfriedersdorf greisen-vein deposit, samples E-148 and E-199, Sauberg mine, underground level 6; (3) Altenberg Sn porphyry, Germany, samples ALT-1 and ALT-2, underground exposures at Sammelrolle 703 and Strecke 3274; (4) Cinovec-Zinnwald Li-Sn greisen or albite granite, sample ZW-1, high-grade greisenized albite granite from an underground exposure (Mineral Collections Natural History Museum London, BM 1991,69); and (5) Krupka Sn greisen, sample KP-1 from the open pit. More detailed deposit and sample characterizations are in the GSA Data Repository[1]. U-Pb dating was carried out using a laser ablation–inductively coupled plasma–mass spectrometry (LA-ICP-MS) system, which consists of an Agilent 7900 quadrupole ICP-MS coupled with a Resonetics RESOlution S-155 laser. See the GSA Data Repository for a full description of the method and error discussion.

RESULTS

All cassiterite samples have high Ti and W, but low Nb and Ta contents, as reflected in their distinct cathodoluminescence oscillatory zoning (Fig. 2), and only a few grains are crosscut by cassiterite stringers, indicating that most of the cassiterite grains grew in one stage. The samples have total Pb contents from 0.01 ppm to 2.27 ppm and low Th contents of <0.23 ppm; some analyses have relatively high common Pb and low concordances. The U contents vary from 0.13 ppm to 37.2 ppm, with a similar variation range in all five deposits.

The cassiterite U-Pb Wetherill concordia ages, Tera-Wasserburg intercepts, and weighted means are in good agreement for the individual samples and define an age spectrum of 326–320 Ma with an uncertainty of ~3 m.y. (Table 1; see $^{207}Pb/^{235}U$ versus $^{206}Pb/^{238}U$ and $^{238}U/^{206}Pb$ versus $^{207}Pb/^{206}Pb$ plots in the Data Repository). The concordia ages and weighted $^{206}Pb/^{238}U$ ages are based on data with ≥95% concordance, which represents ~60% of all measurements. We interpret these data as the cassiterite crystallization age, i.e., the timing of the Sn mineralization event in the deposits sampled.

[1]GSA Data Repository item 2017364, deposit and sample characterization, CL images, U-Pb dating method and data processing, and analytical data, is available online at http://www.geosociety.org/datarepository/2017/ or on request from editing@geosociety.org.

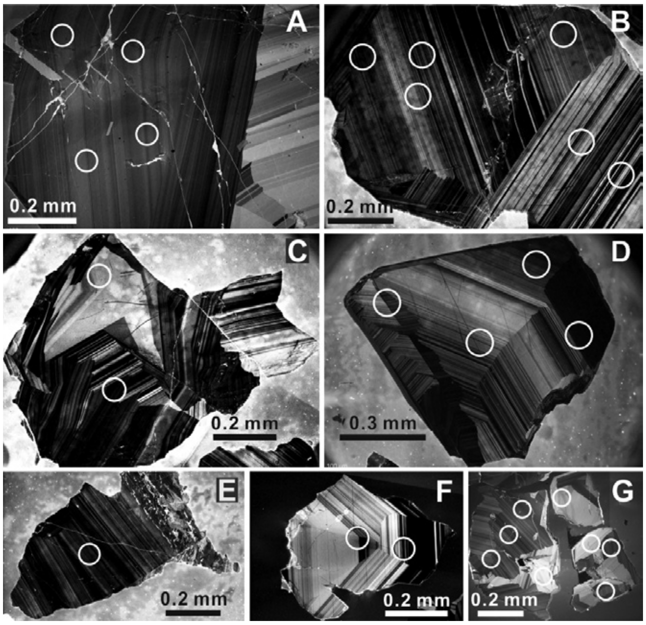

Figure 2. Cathodoluminescence images of representative cassiterite samples analyzed by laser ablation–inductively coupled plasma–mass spectrometry (LA-ICP-MS). The spot size of the laser beam is 74 µm (white circles). Details are in the Data Repository (see footnote 1). A: Sample SD-1 from Sadisdorf with distinct oscillatory zoning cut by white late-stage stringers. B, C: Samples E-148 and E-199 from Ehrenfriedersdorf. D, E: Samples ALT-1 and ALT-2 from Altenberg. F: Sample ZW-1 from Zinnwald-Cinovec. G: Sample KP-1 from Krupka.

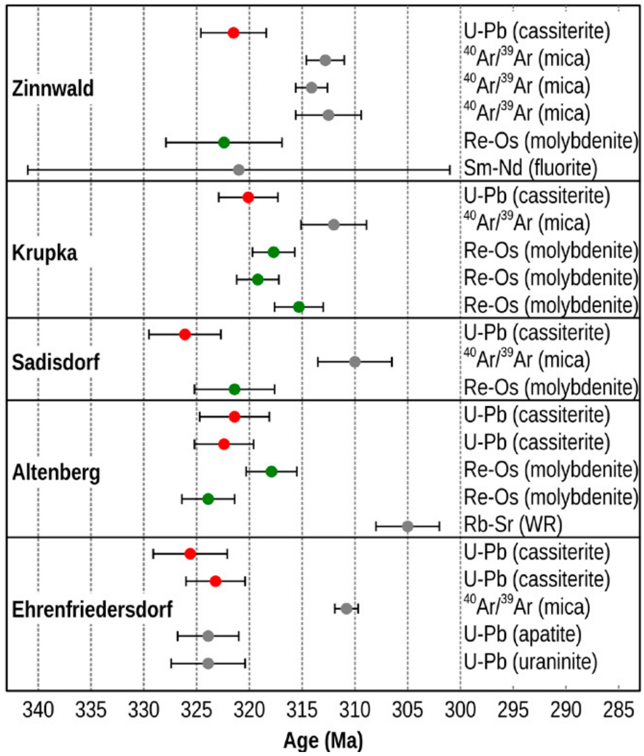

Figure 3. A selection of age data available on the hydrothermal systems studied and their related granites. U-Pb age data on cassiterite in red are from this study. The ^{40}Ar-^{39}Ar age data on mica are from Seifert and Pavlova (2016); Re-Os data on molybdenite are from Romer et al. (2007) and Ackerman et al. (2017); U-Pb ages on uraninite and apatite are from Romer et al. (2007); Rb-Sr whole rock (WR) errorchron age on Altenberg is from Gerstenberger (1989); and Sm-Nd isochron age on fluorite from Zinnwald is from Höhndorf et al. (1994).

DISCUSSION

Previous K/^{40}Ar-^{39}Ar, Rb-Sr, and Sm-Nd age data from the granites of the Erzgebirge-Krušné hory show a large time span, from 330 to 290 Ma (Gerstenberger, 1989; Kempe et al., 2004; Romer et al., 2007; Seifert et al., 2011, Seifert and Pavlova, 2016). ID-TIMS U-Pb dating on zircon from a microgranite dike in the Altenberg area gave apparent ^{206}Pb/^{238}U ages of 366–292 Ma (air abraded), 320–311 Ma (weakly air abraded), and 291–252 Ma (nonabraded), indicating that even TIMS single zircon analysis could not deliver robust U-Pb ages (Romer et al., 2010). Nevertheless, adjusting for minor inheritance and open-system behavior, a Wetherill concordia intercept at 319.2 ± 2.4 Ma (2σ) is currently the best estimate for the age of the alkali feldspar granite magmatism in the Altenberg area (Romer et al., 2010). This age overlaps within uncertainty with our two LA-ICP-MS ages on Altenberg cassiterite (Fig. 3). The Ehrenfriedersdorf tin district is related to a series of highly evolved granite cupolas; the Greifensteine alkali feldspar granite has a U-Pb age of 323.9 ± 3.5 Ma (2σ, mean square of weight deviates = 0.96) for uraninite inclusions in mica (Romer et al., 2007). This age also coincides within uncertainty with our two dates on cassiterite (Fig. 3). The Krasno tin greisen deposit in the Karlovy Vary pluton on the Czech side of the western Erzgebirge-Krušné hory (~50 km south of Ehrenfriedersdorf) was recently dated by the Re-Os method on molybdenite (Ackerman et al., 2017). The age of 323.3 ± 1.6 Ma (2σ) agrees with our U-Pb ages on cassiterite from Ehrenfriedersdorf. Our cassiterite U-Pb ages on Zinnwald-Cinovec and Sadisdorf agree within uncertainty with recent Re-Os model ages on molybdenite of 322.4 ± 5.5 Ma (2σ) and 321.4 ± 3.8 Ma (2σ), respectively (Ackerman et al., 2017).

The available ^{40}Ar-^{39}Ar ages on mica from greisen zones of Zinnwald-Cinovec, Sadisdorf, and Ehrenfriedersdorf consistently show ages in the range of 315–310 Ma (Seifert and Pavlova, 2016) (Fig. 3), i.e., ~10 m.y. after granite emplacement and cassiterite mineralization; this seems to identify a separate time marker for a later regional thermal event. This is supposedly represented by recurrent ignimbrite activity and high heat flow that are documented in the Teplice area and the Erzgebirge molasse basins between 310 and 294 Ma (Hoffmann et al., 2012). Our new and previous

TABLE 1. STATISTICAL DATA FOR THE U-Pb ISOTOPE DATA ON CASSITERITE SAMPLES FROM FIVE TIN DEPOSITS OF THE ERZGEBIRGE PROVINCE

	Wetherill concordia				Tera-Wasserburg				Weighted mean ^{206}Pb/^{238}U		
Locality	**n**	**Age (Ma)**	**2σ**	**MSWD**	**n**	**Age (Ma)**	**2σ**	**MSWD**	**Age (Ma)**	**2σ**	**MSWD**
Sadisdorf (SD-1)	30	326.1	3.4	0.06	73	326.4	2.7	0.50	326.1	3.4	0.30
Ehrenfriedersdorf (E-148)	21	325.6	3.5	0.04	35	325.0	4.0	0.49	325.6	3.5	0.53
Ehrenfriedersdorf (E-199)	32	323.2	2.8	0.04	37	323.5	3.8	0.33	323.2	2.8	0.45
Altenberg (ALT-1)	20	321.4	3.3	0.01	31	320.9	3.3	0.82	321.5	3.3	0.69
Altenberg (ALT-2)	16	322.4	2.8	0.35	19	322.0	2.9	0.47	322.4	2.7	0.49
Zinnwald (ZW-1)	35	321.6	3.1	0.30	71	323.0	2.4	0.69	321.5	3.1	0.21
Krupka (KP-1)	24	320.1	2.8	0.17	36	320.8	2.7	1.14	320.1	2.8	0.97

Note: Concordia ages and weighted mean ^{206}Pb/^{238}U ages are filtered for ≥95% concordance. MSWD—mean square of weighted deviates.

data suggest a relatively narrow time interval of regionally distributed alkali feldspar granite magmatism and associated tin mineralization. The time span of ca. 326–320 Ma would correspond to the model of a large composite granite batholith system underlying the Erzgebirge-Krušné hory region with a long-lasting thermal history (Štemprok and Blecha, 2015). Such a large-scale system would also be able to evolve into highly fractionated small residual melt portions, the alkali feldspar granites, and sustain large-scale fluid flow focused on granite elevations.

CONCLUSIONS

The 20 m.y. hiatus between older biotite granites and the younger Li-mica granites (and their associated Sn deposits) was based on doubtful geochronological data. This problem has now been resolved by bringing field evidence and direct dating of mineralization (Re-Os molybdenite, U-Pb cassiterite) into a new, consolidated context, as the mineralization ages dictate the minimum age for the magmatic crystallization ages. In our study we can reliably show that there is no extended time gap between older and younger granite suites and tin mineralization; all happened within a relatively short time interval of 6.0 ± 4.4 m.y. (326.1 ± 3.4 to 320.1 ± 2.8 Ma).

LA-ICP-MS U-Pb cassiterite dating produces consistent and reproducible dates within single deposits and allows a greater number of analyses from multiple crystals in polished thin sections or slabs, compared with other techniques that require wet chemistry and mineral separation.

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