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Granitic pegmatites of the beryl–columbite subtype in the Tatric Superunit, Western Carpathians, Slovakia: Variscan age determination by in-situ LA–ICP–MS U–Pb dating of columbite-group minerals

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Abstract: Accessory columbite-(Fe) to tantalite-(Fe) from three granitic pegmatites of the Tatric Superunit (Bratislava Massif of the Malé Karpaty Mts., Bojná Massif of the Považský Inovec Mts. and Suchý Massif of the Strážovské Mts.) was used for dating by the in-situ LA–ICP–MS U–Pb method. The columbite–tantalite crystals were sampled from the most fractionated pegmatite dykes of the beryl–columbite subtype situated in the pre-Alpine, Paleozoic crystalline basement of the Tatric Superunit, Western Carpathians (western and central Slovakia). The obtained columbite–tantalite Concordia ages are as follows: 354.5±4.5 Ma (Jezuitské Lesy pegmatite, the Bratislava granite Massif), 360±5.0 Ma (Moravany nad Váhom, Striebornica Ridge pegmatite, the Bojná Massif), and 352±8.5 Ma (Liešťany, Bystrý Hill pegmatite, the Suchý Massif). The columbite–tantalite ages show Mid-Variscan formation of rare-element pegmatites from the Devonian/Carboniferous boundary to Tournaisian stage, which is coeval with the emplacement of cogenetic granites during the main phase of Variscan intracontinental subduction and collision. The obtained columbite–tantalite age interval of rare-element granitic pegmatites of the Tatric Superunit (~360 to 350 Ma) is generally older than the ages of Be- and Li-rich rare-element pegmatites of the Moldanubian Superunit in the Bohemian Massif (~340 to 320 Ma). The rare-element granitic pegmatites of the Austroalpine Superunit (Eastern Alps) are significantly younger (~290 to 240 Ma) because they were formed in an extension regime during the Permian to Early Triassic post-Variscan orogenic collapse.

Keywords: columbite, tantalite, granitic pegmatite, LA–ICP–MS U–Pb dating, Variscan orogeny, Tatric Superunit, Western Carpathians

Introduction

Columbite-group minerals [(Fe,Mn)(Nb,Ta)$_2$O$_6$] are the most common and characteristic accessory minerals of the evolved rare-element class, beryl type, and LCT family granitic pegmatites (e.g., Černý & Ercit 1989, 2005; London 2008, and references therein). They are sensitive indicators of parental pegmatite evolution from magmatic to post-magmatic (hydrothermal, metamorphic) stages. They show primary regular magmatic growth and fractionation trends, generally from columbite-(Fe) to tantalite-(Fe) and -(Mn) (Černý et al. 1986; Černý 1989), as well as secondary alteration and dissolution–reprecipitation internal textures and changes in their chemical composition (e.g., Tindle & Breaks 1998; Novák et al. 2003; Pieczka 2010; Chudík et al. 2011; Melcher et al. 2015; Konzett et al. 2018; Chládek et al. 2020; Shaw et al. 2022; Yuan et al. 2022; Araujo et al. 2023).

Progress in analytical techniques during the last decades enables the application of U–Pb geochronology to columbite-group minerals. They contain a sufficiently high concentration of uranium in its structure for U–Pb dating, usually in the range of ~3 to 10,000 ppm U, rarely up to 3.7 wt.% UO$_2$, as well as the low incorporation of common lead (Romer & Wright 1992; Romer & Smeds 1994, 1997; Smith et al. 2004; Baumgartner et al. 2006; Melleton et al. 2012; Deng et al. 2013; Tang et al. 2017; Legros et al. 2019; Feng et al. 2020; Peytcheva et al. 2021). The first isotopic U–Pb dating were based on the classic method of chemical dissolution of columbite–tantalite concentrate (Romer & Wright 1992; Romer & Smeds 1994, 1997; Smith et al. 2004; Baumgartner et al. 2006). Since the 2010s, the in situ LA–ICP–MS or SIMS techniques have represented the main methods for obtaining

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the U–Pb isotopic age of columbite–tantalite (e.g., Melleton et al. 2012; Deng et al. 2013; Konzett et al. 2018; Legros et al. 2019; Galliski et al. 2021, Peycheva et al. 2021; Yuan et al. 2022). It is important to obtain geochronological data on the pegmatites for better constraining of the geological processes at their origin, their mutual genetic relationships to adjacent granitic rocks, or metamorphic processes. Moreover, the rare-element granitic pegmatites represent valuable mineral resources, especially of critical metals (including Nb and Ta) or industrial minerals, and knowledge of their age relationships allow us to develop an effective exploration strategy of searching the potential deposits of the critical metals.

In this contribution, we present the first in situ LA–ICP–MS U–Pb dating of columbite-group minerals from the three granitic pegmatites of the beryl–columbite subtype in the Tatric Superunit, Western Carpathians (Slovakia). Moreover, the paper provides a compositional characterization of the investigated columbite–tantalite crystals and their age comparison with parental granites, as well as rare-element pegmatites in the adjacent terranes of Central Europe. The presented ages are the first application of the U–Pb geochronology on Variscan Nb–Ta oxide minerals in the entire Carpathian arc.

Regional geology

The investigated granitic pegmatites, together with adjacent granitic and metamorphic rocks, are a part of Paleozoic (Variscan) crystalline basement of the Tatric Superunit in the Central Western Carpathians (Fig. 1). The Variscan West-Carpathian crystalline basement was incorporated into an Alpine nappe tectonic structure, which is a part of the Alps–Carpathians Mountain range, formed during the Mesozoic to Cenozoic Alpine orogenic evolution (e.g., Plašienka 2018). The pegmatite parental granitic rocks represent biotite to biotite–muscovite granites, granodiorites, rarely (leuco)tonalites, locally with entrapped small diorite bodies. The host granitic rocks show an orogen-related, peraluminous calc-alkaline character with S- to I-type geochemical affinity originating in a subduction- to collision-related tectonic regime (e.g., Broska & Uher 2001; Kohút & Larionov 2021). Locally, more evolved muscovite–(biotite) leucogranites, pegmatitic leucogranites, aplites, and pegmatites occur in apophyses and/or dyke bodies within the granitic massifs and are hosted by metamorphic rocks. The granitic rocks show a Variscan age of magmatic crystallization, which are usually in an interval from around 360 to 340 Ma, based on numerous multi-grain zircon U–Pb, whole rock and mica Rb–Sr, and K–Ar isotopic data (summarized in Cambel et al. 1990), as well as on in-situ zircon U–Pb and Pb–Pb isotopic dating (e.g., Kohút & Larionov 2021, and references therein; Broska et al. 2022) and monazite in-situ Th–U–Pb EPMA chemical dating (Finger et al. 2003; Uher et al. 2014).

The metamorphic rocks of the Tatric Superunit basement represent Lower Paleozoic (usually Ordovician to Devonian) metapelites to metapsammites, typically of the greenschist to amphibolite facies, mainly phyllites, biotite-, garnet-, staurolite- and sillimanite-bearing micaschists to paragneisses, less frequently orthogneisses (metagranites) and amphibolites (metagabbros to metabasaltic rocks), rarely metacarbonates, metaquartzites, carbon-rich metapelites (black schists), and serpentinized metaultrabasic rocks (e.g., Krist et al. 1992, and references therein).

The pegmatites form tabular bodies (dykes) and less frequently, lensoid bodies, which had intruded into the granitic rocks, rarely into the adjacent metamorphic rocks in close exocontact with the granite intrusions. The thickness of the pegmatites is usually from 0.5 to 2 m, and locally up to 8 m (the Moravany nad Váhom, Striebornica Ridge pegmatite), and they attain a length of ~100 m; the contacts with adjacent rocks are usually sharp. The pegmatites commonly show internal, more or less well-developed zoning, generally with a fine-crystalline aplitic to coarse-crystalline quartz + K-feldspar (microcline) + albite + muscovite ± biotite (annite) border to an intermediate unit, an intermediate zone with coarse-grained to blocky K-feldspar (± quartz, muscovite, or biotite), as well as a quartz core zone. Locally, irregular late albite-rich assemblages with dominant fine-grained saccharoidal or platy albite (cleavelandite variety) aggregates partially replaced the older pegmatite units. Accessory minerals include predominantly garnet (almandine–spessartine), less abundant metamict Hf-rich zircon, fluorapatite, rarely gahnite, monazite-(Ce), xenotime-(Y), uraninite, pyrite, native bismuth, and other less frequent phases.

The granitic pegmatites with beryl and Nb–Ta oxide minerals represent the most evolved pegmatite population related to the Variscan granitic massifs of the Tatric Superunit. They occur mainly in the Bratislava Massif of the Malé Karpaty Mts., less frequently in other areas, such as the Bojná Massif of the Považský Inovec Mts., Žiar Mts., Suchý Massif of the Strážovské Mts., and the Nízke and Vysoké Tatry Mts. Beryl forms 2 to 5 cm, exceptionally up to ca. 20 cm, long prismatic hexagonal crystals with moderately elevated contents of Fe, Mg, and Na; Cs and Li are exceptionally high (Uher et al. 2010; Bačík et al. 2019). Beryl is frequently altered to secondary post-magmatic phenakite and bertrandite (Uher et al. 2022).

The primary magmatic accessory Nb–Ta phases comprise predominantly columbite-group minerals [columbite-(Fe), columbite-(Mn), tantalite-(Fe), and tantalite-(Mn)], rarely tapiolite-(Fe), ferrowodginite, Ti-rich ixiolite, Nb–Ta-rich rutile, which are commonly associated with secondary Nb–Ta minerals, such as fersmite, pyrochlore-supergroup minerals (mainly members of microlite-group), and locally Nb–Ta-rich titanite (e.g., Uher et al. 1994, 1998a, b, 2007, 2020; Uher & Broska 1995; Novák et al. 2000; Chudík et al. 2011). The columbite-group minerals and other Nb–Ta oxide phases are usually in association with muscovite, quartz, K-feldspar, albite, almandine–spessartine, and beryl in intermediate to core zones or saccharoidal to cleavelandite albite-rich replacement units.

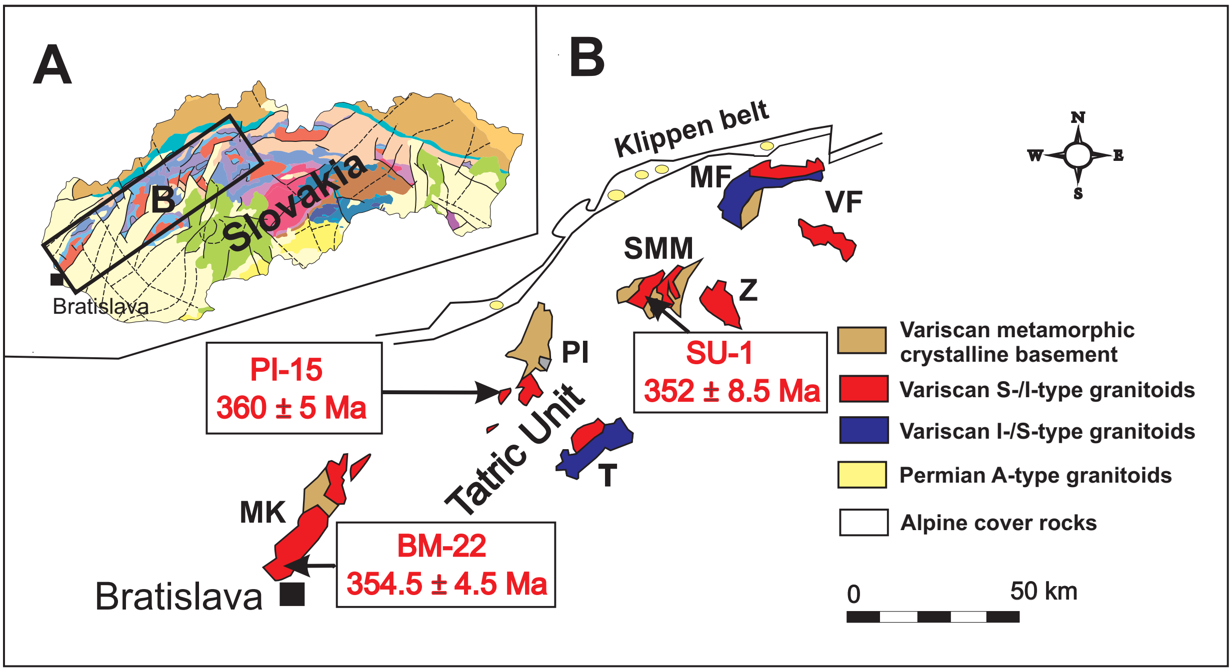

Fig. 1. Simplified geological map with location and corresponding columbite–tantalite U–Pb ages from the investigated granitic pegmatites in the Tatric Superunit of the Western Carpathians.

The mineral association and its chemical composition, as well as the geochemical signatures of the most evolved granitic pegmatites of the Tatric Superunit enable the classification of them into the lithium–cesium–tantalum (LCT) family, beryl type, and beryl–columbite subtype of the rare-element class (Černý & Ercit 2005).

Materials and methods

Columbite–tantalite samples

For the purpose of in-situ U–Pb dating, we separated columbite–tantalite crystals from three typical samples of the most evolved granitic pegmatites in the Tatric Superunit: (1) Bratislava, Jezuitské Lesy in the Bratislava Massif, the Malé Karpaty Mountains, (2) Moravany and Váhom, Striebornica Ridge in the Bojná Massif, the Považský Inovec Mts., and (3) Liešťany, Bystrý Hill, in the Suchý Massif, the Strážovské Mts. (Fig. 1). For dating and analytical purposes, unaltered, black euhedral tabular crystals of columbite–tantalite with a metallic luster of ~0.5 to 5 mm in size were selected. The studied columbite–tantalite is associated with saccharoidal albite, quartz, muscovite, and almandine–spessartine in albitized coarse-crystalline quartz+K-feldspar (microcline)+muscovite zone of the pegmatites. Detailed information on analytical data concerning the mineralogy and geology of the investigated pegmatites have been previously published (e.g., Uher et al. 1994, 2010, 2020, 2022; Novák et al. 2000; Chudík et al. 2011).

Electron probe microanalysis (EPMA)

The chemical composition of the separated columbite–tantalite crystals was measured on polished sections (2.5 cm in diameter) using the JEOL JXA–8530F field-emission electron-probe microanalyser in the wave-dispersion mode (WDS) at the Earth Science Institute of the Slovak Academy of Sciences in Banská Bystrica, Slovakia. The following analytical conditions were used: accelerating voltage of 15 kV, a probe current of 20 nA, beam diameter of 1–3 µm, 20 seconds counting time at the peak for the measured elements (30 s for Th, U and Pb), and ZAF matrix correction was used. The following calibrants, X-ray lines, and detection limits (3σ, in ppm) were used: scheelite (W Lα, 640), $LiNbO_3$ (Nb Lα, 380), $CrTa_2O_6$ (Ta Mα, 500), rutile (Ti Kα, 490), ZrO_2 (Zr Lα, 230), cassiterite (Sn Lα, 160), ThO_2 (Th Mα, 180), UO_2 (U Mβ, 270), $ScVO_4$ (Sc Kα, 150), YPO_4 (Y Lα, 270), $CePO_4$ (Ce Lα, 920), stibnite (Sb Lα, 190), BiTe (Bi Lα, 650), hematite (Fe Kα, 350), rhodonite (Mn Kα, 370), willemite (Zn Kα, 770), diopside (Mg Kα, 100; Ca Kα, 80), and crocoite (Pb Mβ, 340). The same electron-probe apparatus was used for a detailed study of mineral textural relations, with an application of back-scattered electron (BSE) images.

LA–ICP–MS U–Pb dating

The laser ablation inductively-coupled plasma mass spectrometry (LA–ICP–MS) method was applied for in situ U–Pb dating of columbite–tantalite crystals. Only primary, relatively large and homogeneous parts of columbite-(Fe) or tantalite-(Fe) crystals showing regular growth zoning or unzonal internal texture without late irregular recrystallization patterns under BSE were selected for the following in-situ LA–ICP–MS U–Pb dating. Analytical work was performed using the equipment at the Geological Institute of the Bulgarian Academy of Sciences, Sofia: the NWR UP193 FX laser ablation system combined with the PerkinElmer ELAN DRC-e ICP-mass spectrometer. The laser pit was 35 μm diameter, with laser beam repetition rate of 4 Hz and energy density of 4.6–4.7 j/cm^2 at the sample surface. The operating conditions, data acquisition parameters, measured isotopes, and standardization are disscussed in detail in Peycheva et al. (2021). The X36 columbite with an age of 354.8 ± 1.7 Ma (Galliski et al. 2021) was used as the primary matrix-matched reference material. The data reduction was done in Iolite version (Paton et al. 2011) combined with the VisualAge (Petrus & Kamber 2012) to obtain ratios corrected for instrumental fractionation and drift and down-hole fractionation. The geochronological plots and ages were produced in ISOPLOT 3.75 program (Ludwig 2012). Analytical conditions are summarized in Appendix 1.

Results

Columbite–tantalite chemical composition and zoning

The electron probe microanalysis (EPMA) and BSE images documented the compositional zoning and chemistry of the investigated columbite–tantalite crystals (Table 1, Figs. 2 and 3). Two investigated crystals from the Bratislava, Jezuitské Lesy pegmatite show relatively homogeneous internal texture, locally with irregular zones and with a slightly lower Ta/(Ta+Nb) atomic ratio. All measured EPMA spots showed tantalite-(Fe) composition with Mn/(Mn+Fe)=0.42–0.47 and Ta/(Ta+Nb)=0.59–0.69 (Fig. 3). The analyzed columbite-group minerals from the Moravany nad Váhom pegmatite revealed crystals with relatively homogeneous zones, but also with distinct regular magmatic growth zoning and locally with late irregular dissolution–reprecipitation rim domains as well (Fig. 2). The primary zones show columbite-(Fe) composition with Mn/(Mn+Fe)=0.19–0.33 and Ta/(Ta+Nb)=0.19–0.41, whereas late irregular domains reveal different columbite-(Fe) to tantalite-(Fe) composition with relatively narrow Mn/Fe, but a very broad Ta/Nb ratio, where Mn/(Mn+Fe)=0.15–0.21 and Ta/(Ta+Nb)=0.07–0.61 (Table 1, Fig. 3). One investigated crystal from the Liešťany pegmatite represents a relatively homogeneous columbite-(Fe) crystal with a relatively homogeneous chemical composition [Mn/(Mn+Fe)=0.38–0.40, Ta/(Ta+Nb)=0.21–0.24], but contains narrow irregular replacement rims of Ta-rich columbite-(Fe) to tantalite-(Fe) with Mn/(Mn+Fe)=0.40–0.41 and Ta/(Ta+Nb)=0.45–0.62 (Table 1, Figs. 2 and 3).

The contents of Ti, W, Sn, Zr, Sn, U, and other elements measured by EPMA are generally low for all of the investigated columbite–tantalite crystals used for the U–Pb dating; they usually attained less than ~1 wt. % of oxide. The uranium contents measured in the studied columbite–tantalite crystals by the LA-ICP-MS method attain 10 to 50 ppm in the Bratislava, Jezuitské Lesy, 13 to 162 ppm in the Moravany nad Váhom, Striebornica Ridge, and 10 to 16 ppm in the Liešťany, Bystrý Hill pegmatite (Appendix 2).

U–Pb dating of columbite–tantalite

In-situ LA–ICP–MS U–Pb isotopic dating of selected zones of columbite–tantalite crystals from the investigated granitic pegmatites gave the following average concordia $^{206}Pb/^{238}U$ ages (Appendix 2, Figs. 2 and 4):

- 354.5±4.5 Ma for the Bratislava, Jezuitské Lesy pegmatite (BM-22; Bratislava Massif, the Malé Karpaty Mts.),
- 360±4.5 Ma for the Moravany and Váhom, Striebornica Ridge pegmatite (PI-15; Bojná Massif, the Považský Inovec Mts.), and
- 352±8.5 Ma for the Liešťany, Bystrý Hill pegmatite (SU-1; Suchý Massif, the Strážovské Mts.).

The obtained ages are slightly discordant (mainly the Jezuitské Lesy and Liešťany pegmatites), but the analyzed points are situated near the ideal Concordia line. Moreover, several measured LA–ICP–MS spots yielded discordant, apparently older (376 to 1124 Ma) or younger (129 to 294 Ma) $^{206}Pb/^{238}U$ ages in all of the three columbite–tantalite samples.

Discussion

Columbite–tantalite age results

Our analytical results represent the first application of the U–Pb geochronology based on Nb–Ta oxide minerals in the Western Carpathian area, as well as in the entire Carpathian mountain arc. The in-situ LA–ICP–MS U–Pb dating of columbite-(Fe) and tantalite-(Fe) from three granitic pegmatites hosted in the Variscan Tatric crystalline basement of the Western Carpathians reveal their Mid-Variscan age range of 360±4.5 to 352±8.5 Ma, from the Devonian/Carboniferous boundary to the Tournaisian stage (according to the International Chronostratigraphic Chart 2023, Version 09; www.stratigraphy.org). The discordant ages, which are apparently older (376 to 1124 Ma) or younger (129 to 294 Ma) $^{206}Pb/^{238}U$ in all three columbite–tantalite samples, most likely have no geologically signficant meaning. Such ages are usually connected with damaged domains in the mineral structure due to its metamictization and subsequent Pb-loss, or the presence of nanoscale uraninite or other U–(Th) mineral inclusions, which are not visible in BSE images. The problem and possible origin of such obtained inconsistent columbite–tantalite ages has been previously discussed in several studies (e.g., Romer et al. 1996; Romer 2003; Smith et al. 2004; Legros et al. 2019).

Table 1: Electron-probe microanalyses of columbite–tantalite from investigated granitic pegmatites (oxides in wt. %).

Location	Bratislava, Jezuitské Lesy		Moravany nad Váhom, Striebornica Ridge				Lieštany, Bystrý Hill	
Crystal/Site	Cr2/cen	Cr2/cen	Cr1/rim	Cr1/repl	Cr2/rim	Cr5/repl	Cr1/cen	Cr1/repl
Analysis #	BM-22-1B5	BM-22-1B7	PI-15-2.18	PI-15-2.20	PI-15-2.22	PI-15-2.30	SU-1.9	SU-1.12
Mineral	Tan-Fe	Tan-Fe	Tan-Fe	Col-Fe	Col-Fe	Tan-Fe	Col-Fe	Tan-Fe
WO_3	0.00	0.53	0.35	0.00	0.09	0.45	0.18	0.38
Nb_2O_5	19.50	17.51	23.18	69.85	40.69	27.16	56.28	21.99
Ta_2O_5	64.21	63.53	58.95	8.65	40.51	53.60	24.33	60.44
TiO_2	0.09	0.23	0.51	0.62	0.52	1.14	0.26	0.21
ZrO_2	0.00	0.03	0.16	0.03	0.08	0.30	0.03	0.00
SnO_2	0.17	0.10	0.03	0.00	0.06	0.10	0.11	0.04
UO_2	0.00	0.00	0.00	0.00	0.07	0.09	0.00	0.07
Sc_2O_3	0.06	0.00	0.07	0.00	0.00	0.05	0.00	0.05
Bi_2O_3	0.00	0.18	0.13	0.00	0.00	0.00	0.00	0.00
Fe_2O_3	0.15	2.55	1.69	0.85	1.26	2.42	0.79	2.28
FeO	8.94	6.52	11.93	16.20	13.44	11.47	11.31	7.93
MnO	6.59	7.44	3.12	3.53	3.51	3.57	7.35	6.82
ZnO	0.00	0.00	0.00	0.00	0.00	0.00	0.13	0.00
MgO	0.00	0.00	0.03	0.07	0.03	0.00	0.00	0.00
Total	99.71	98.62	100.15	99.80	100.26	100.35	100.77	100.21
Mineral formulae based on 6 O atoms, 3 cations and valence calculation								
W	0.000	0.010	0.007	0.000	0.002	0.008	0.003	0.007
Nb	0.667	0.605	0.765	1.832	1.223	0.867	1.566	0.730
Ta	1.322	1.320	1.170	0.137	0.733	1.029	0.407	1.208
Ti	0.005	0.013	0.028	0.027	0.026	0.061	0.012	0.012
Zr	0.000	0.001	0.006	0.001	0.003	0.010	0.001	0.000
Sn	0.005	0.003	0.001	0.000	0.002	0.003	0.003	0.001
U	0.000	0.000	0.000	0.000	0.001	0.001	0.000	0.001
Sc	0.004	0.000	0.004	0.000	0.000	0.003	0.000	0.003
Bi	0.000	0.004	0.002	0.000	0.000	0.000	0.000	0.000
Fe^{3+}	0.008	0.147	0.093	0.037	0.063	0.128	0.037	0.126
Fe^{2+}	0.566	0.416	0.728	0.786	0.747	0.677	0.582	0.487
Mn	0.423	0.481	0.193	0.174	0.198	0.213	0.383	0.424
Zn	0.000	0.000	0.000	0.000	0.000	0.000	0.006	0.000
Mg	0.000	0.000	0.003	0.006	0.003	0.000	0.000	0.000
Cation sum	3.000	3.000	3.000	3.000	3.001	3.000	3.000	2.999
Mn/(Mn + Fe)	0.42	0.46	0.19	0.17	0.20	0.21	0.38	0.41
Ta/(Ta + Nb)	0.66	0.69	0.60	0.07	0.37	0.54	0.21	0.62

Th, V, Y, Sb, Ca and Pb contents are below detection limit.
Cr = crystal, cen = central part od crystal, rim = rim part of crystal; repl = replacement domain,
Tan-Fe (tantalite-(Fe)), Col-Fe (columbite-(Fe))
Values of Fe^{3+} and Fe^{2+} (and corresponding Fe_2O_3 and FeO) were calculated on assumption
of neutral charge balance where cation charge = +12 and anion charge (6 O^{2-}) = −12

Ages of pegmatite columbite–tantalite and related Variscan granites

The obtained 360±4.5 to 352±8.5 Ma age interval of the investigated columbite-group minerals is consistent with the magmatic crystallization and emplacement age of the hosted and adjacent Variscan S-type (or mixed S/I-type) granitic rocks from the Bratislava, Bojná, and Suchý & Malá Magura massifs and determined repeatedly by U–Pb method on zircon (~360 to 350 Ma, Kráľ et al. 1997; Kohút et al. 2009; Kohút & Larionov 2021; Table 2). The columbite–tantalite age from the Bratislava, Jezuitské Lesy pegmatite (354.5±4.5 Ma) is almost identical with a SHRIMP U–Pb age of zircon from the adjacent biotite granodiorite of the Bratislava Massif, located in Bratislava, Devín quarry (355.4±4.7 Ma, Kohút et al. 2009), approximately 2 km SW of the investigated pegmatite dyke. The columbite–tantalite age from the Moravany nad Váhom, Striebornica Ridge pegmatite, Považský Inovec Mts. shows the oldest age among the investigated three samples (360±4.5 Ma), in accordance with age obtained from adjacent granites, which also show a slightly older, Upper Devonian age (367±5 Ma; Kohút & Larionov 2021). The age of columbite from the Lieštany, Bystrý Hill pegmatite in the Suchý Massif, Strážovské Mts. (352±8.5 Ma) fits well with the dating of adjacent Variscan granites with a zircon Pb–Pb TIMS age at 356±9 Ma (Kráľ et al. 1997). Moreover, our columbite–tantalite dating shows very good agreement with other age determinations made by the monazite Th–U–Pb

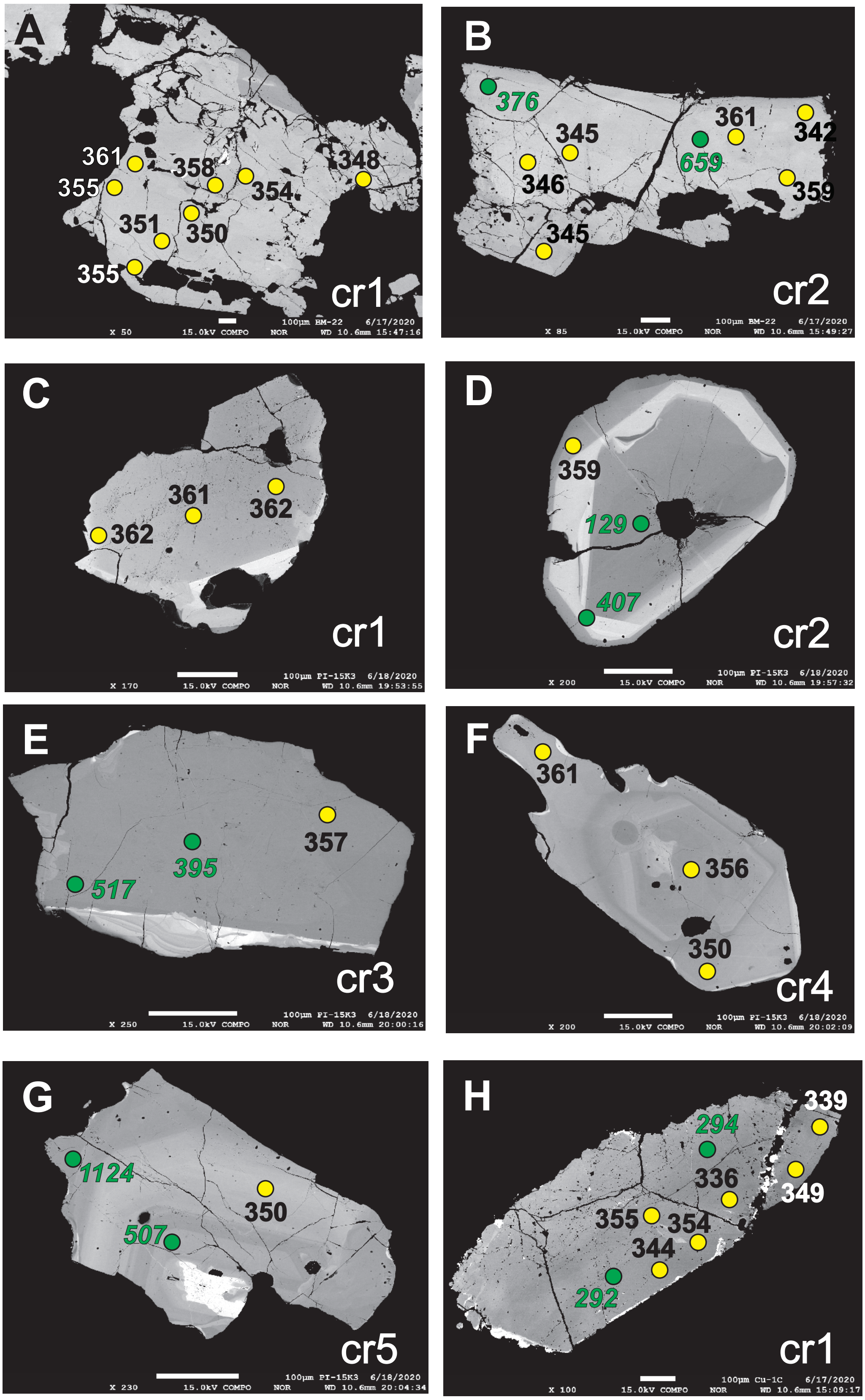

Fig. 2. Back-scattered electron images of the studied columbite–tantalite with locations of dated spots and corresponding $^{206}Pb/^{238}U$ ages (in Ma). **A–B**: Bratislava, Jezuitské Lesy pegmatite (BM-22 sample), crystals 1 and 2 (cr1, cr2); **C–G**: Moravany nad Váhom, Striebornica Ridge pegmatite (PI-15 sample), crystals 1 to 5 (cr1 to cr5); **H**: Liešťany, Bystrý Hill pegmatite (SU-1 sample), crystal 1 (cr1). Yellow spots = concordant Variscan ages, green spots = discordant non-Variscan ages.

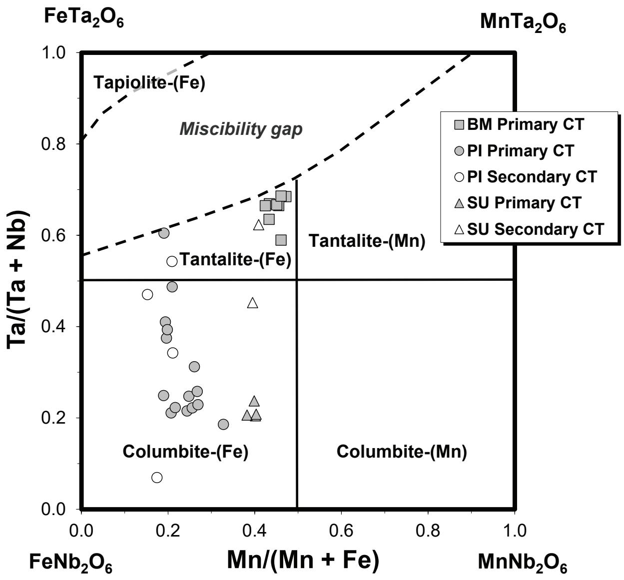

Fig. 3. Quadrilateral Ta/(Ta+Nb) versus Mn/(Mn+Fe) diagram (atomic ratios) of primary and secondary columbite–tantalite (CT) illustrating the chemical composition of the investigated crystals of columbite-(Fe) and tantalite-(Fe). BM: Bratislava, Jezuitské Lesy pegmatite; PI: Moravany nad Váhom, Striebornica Ridge pegmatite; SU: Liešťany, Bystrý Hill pegmatite.

EPMA and molybdenite Re–Os N–TIMS methods on adjacent granites and pegmatites (Finger et al. 2003; Putiš et al. 2009; Uher et al. 2014; Kohút & Larionov 2021; Table 2).

The synchronous ages of Variscan pegmatites and host granitic rocks, as well as the prevailing position of the pegmatite dykes within the granitic plutons, whole-rock petrography, geochemistry, and mineral composition of the granites with the presence of transitional pegmatitic and aplitic leucogranites (e.g., Cambel & Vilinovič 1987; Dávidová 1994, 1997; Uher & Broska 1995; Uher & Černý 1998; Uher et al. 2010, 2022) confirm the parental genetic relationship of the granitic magma to the fractionated pegmatite melt in the Tatric Superunit.

Formation of rare-element granitic pegmatites in the frame of Variscan to post-Variscan Central Europe

The determined columbite–tantalite age interval of the Tatric rare-element granitic pegmatites is contemporaneous with huge, orogen-related, calc-alkaline granitic magmatism in the wide collision zone between the Gondwana and Laurussia continents during the final intracontinental subduction stage of Variscan orogeny, taking place approximately 360 to 340 Ma ago (e.g., Kroner & Romer 2013, and references therein). Variscan beryl-type, columbite subtype granitic pegmatites of the Tatric Superunit (~360 to 350 Ma) show an older age interval in comparison with the adjacent pre-Alpine terranes in the Moldanubian (Bohemian Massif) and Austroalpine (Eastern Alps) crystalline basement (Table 3). The rare-element granitic

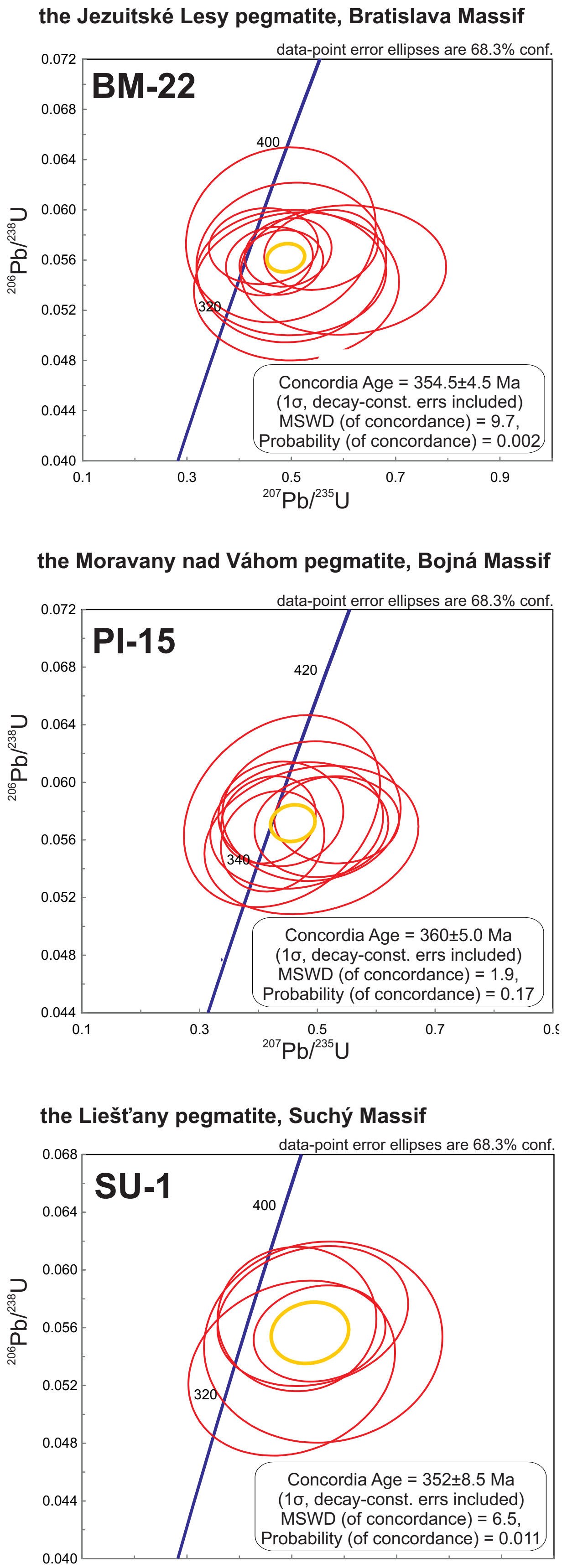

Fig. 4. Concordia diagrams of columbite–tantalite LA–ICP–MS U–Pb dating.

Table 2: Zircon, monazite, columbite–tantalite and molybdenite dating from Variscan granites and pegmatites of the Bratislava, Bojná and Suchý & Malá Magura massifs.

Rock	Mineral, method	Age (Ma) Bratislava Massif	Bojná Massif	Suchý & Malá Magura M.
Granite	Zrn, U–Pb SHRIMP	355±5[a] to 343±2[b]	367±5[c]	361±3[c]
Granite	Zrn, Pb–Pb TIMS			356±9[d]
Granite	Mnz, Th–U–Pb EPMA	359±9[e] to 346±10[e]	364±17[f]	342±13[f]
Pegmatite	Col, U–Pb LA–ICP–MS	354±5*	360±5*	352±9*
Pegmatite	Mol, Re–Os N-TIMS	357±3[c]		
Pegmatite	Mnz, Th–U–Pb EPMA	357±6[e] to 352±5[e]		

References:
* This study
[a] Kohút et al. (2009)
[b] Putiš et al. (2009)
[c] Kohút & Larionov (2021)
[d] Kráľ et al. (1997)
[e] Uher et al. (2014)
[f] Finger et al. (2003)

Mineral abbreviations:
Zrn=zircon
Mnz=monazite
Col=columbite–tantalite
Mol=molybdenite

pegmatites of the Moldanubian Superunit revealed a younger age interval of ~340 to 335 Ma for beryl-type (beryl–columbite±phosphate subtypes) pegmatites (Novák et al. 1998; Novák 2005), and ~340 to 320 Ma for the Li-rich, complex type (elbaite, lepidolite and petalite subtypes) pegmatites (Ertl et al. 2004; Melleton et al. 2012). Rare-element granitic pegmatites of the Austroalpine Superunit are apparently younger; they show a post-Variscan (Permian to Early Triassic) age span of formation: ~290 to 240 Ma for beryl-type, beryl–columbite subtype (Konzett et al. 2018), and ~270 to 240 Ma for Li-rich albite–spodumene type (Thöni & Miller 2000; Knoll et al. 2018).

As a consequence, the populations of rare-element granitic pegmatites in pre-Alpine Central European terranes are not synchronous, and they are closely related with regional tectonic, magmatic, and metamorphic evolution during Variscan orogeny, as well as post-Variscan orogenic collapse and extension during the break-up of the Pangea supercontinent. The Tatric and Moldanubian Be- and Li-rich pegmatites originated synchronously with their more or less related (±parental) granite suites with S-type (or mixed S/I-type) affinity during the main Carboniferous stages of Variscan orogeny, as documented by numerous datings of adjacent granitic rocks (e.g., Žák et al. 2011; Trubač et al. 2017; Janoušek 2019; Kohút & Larionov 2021, and references therein).

Conversely, the rare-element (Be and Li) pegmatites of the Austroalpine Superunit are connected with a post-Variscan, Permian to Early Triassic extensional stage, and they are coeval and related with small intrusions of S-type leucogranites, enriched in Li and Sn (Knoll et al. 2018, 2023). Possibly analogous, Permian S-type leucogranites (~270 to 250 Ma), locally with Li–Sn–Nb–Ta–W mineralization occur in the Gemeric Superunit in the Western Carpathians as well (e.g., Finger & Broska 1999; Broska & Kubiš 2018; Villaseñor et al. 2021), rarely with occurrences of slightly Li-enriched granitic pegmatites, which contain schorl-elbaite, columbite, and cassiterite (Dobšiná area; Uher et al. in prep.). In contrast to the Tatric and Moldanubian rare-element granitic pegmatites, the Austroalpine post-Variscan rare-element granitic pegmatites are recently considered as direct anatectic products of fertile Li-rich metapelite partial melting (Konzett et al. 2018; Knoll et al. 2023).

Conclusions

- Rare-element granitic pegmatites of the beryl type (beryl–columbite subtype) of the Tatric Superunit (Western Carpathians) contain accessory minerals of the columbite group (columbite–tantalite) suitable for geochronological dating in-situ by the LA–ICP–MS U–Pb method.
- The columbite–tantalite dating reveals 360 to 350 Ma age of the magmatic solidification of the pegmatites, coeval with parental Variscan granitic rocks during the main stage of mid-Variscan subduction to collision.
- The Be-rich rare-element granitic pegmatites of the Tatric Superunit are older in comparison with Be- and Li-rich (beryl and complex type), orogen-related Variscan pegmatite populations in the Moldanubian Superunit (Bohemian Massif). The beryl and albite–spodumene (Li-rich) granitic pegmatites of the Austroalpine Superunit (Eastern Alps) are significantly younger, coeval to the post-Variscan orogenic collapse, and extension (Permian to Early Triassic).

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Table 3: Age determinations of rare-element, LCT-family granitic pegmatites of Tatric Unit and related terranes during Variscan (Devonian–Carboniferous) to post-Variscan (Permian) stage.

Geological unit	Locality or area	Pegmatite type	Age (Ma)	Mineral, method
Tatric (W. Carpathians)	Bratislava	Beryl	354±5*	Col, U–Pb LA–ICP–MS
	Moravany nad Váhom	Beryl	360±5*	Col, U–Pb LA–ICP–MS
	Liešťany	Beryl	352±9*	Col, U–Pb LA–ICP–MS
Moldanubian (Bohemian Massif)	Písek	Beryl	339±3[a,b]	Mnz, U–Pb M
	Věžná	Beryl	335±3[a,b]	Mnz, U–Pb M
	Eibenstein an der Thaya	Complex	337±5[c]	Gar–Ab, Sm–Nd M
	Rožná	Complex	332±3[d]	Col, U–Pb LA–ICP–MS
	Dobrá Voda	Complex	332±3[d]	Col, U–Pb LA–ICP–MS
	Dobrá Voda	Complex	328±5[d]	Col, U–Pb LA–ICP–MS
	Puklice	Complex	336±3[d]	Col, U–Pb LA–ICP–MS
	Jeclov	Complex	333±7[d]	Col, U–Pb LA–ICP–MS
	Sedlatice	Complex	334±6[d]	Col, U–Pb LA–ICP–MS
	Chvalovice	Complex	332±3[d]	Col, U–Pb LA–ICP–MS
	Nová Ves	Complex	327±3[d]	Col, U–Pb LA–ICP–MS
	Nová Ves	Complex	323±3[d]	Col, U–Pb LA–ICP–MS
	Ctidružice	Complex	323±5[d]	Col, U–Pb LA–ICP–MS
Austroalpine (Eastern Alps)	Monte Altacroce	Beryl	289±5[e]	Cst, U–Pb LA–ICP–MS
	Monte Altacroce	Beryl	242±8[e]	Ttn, U–Pb LA–ICP–MS
	Monte Altacroce	Beryl	224±12[e]	Ttn, U–Pb LA–ICP–MS
	Hohenwart	Ab-Spd	261±13[f]	WR–Tur–Gar, Sm–Nd M
	Lachtal	Ab-Spd	268±2[f]	WR–Gar, Sm–Nd M
	Mitterberg	Ab-Spd	263±9[f]	WR–Tur–Gar, Sm–Nd M
	Mitterberg	Ab-Spd	272±3[f]	Spd–Kfs, Rb–Sr M
	Mitterberg	Ab-Spd	247±3[f]	Ms–Kfs, Rb–Sr M
	Weinebene	Ab-Spd	243±2[g]	WR–Spd, Rb–Sr M
	Weinebene	Ab-Spd	239±3[g]	WR–Spd, Rb–Sr M

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* This study
[a] Novák et al. (1998)
[b] Novák (2005)
[c] Ertl et al. (2004)
[d] Melleton et al. (2012)
[e] Konzett et al. (2018)
[f] Knoll et al. (2018)
[g] Thöni & Miller (2000)

Abbreviations:
M=multigrain method; Cst=cassiterite
WR=whole rock; Ttn=titanite
Col=columbite–tantalite; Tur=tourmnaline
Mnz=monazite; Spd=spodumene
Gar=garnet; Kfs=K-feldspar
Ab=albite; Ms=muscovite

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Appendix 1

Analytical conditions for LA–ICP–MS U–Pb dating of columbite–tantalite.

ICP–MS	PerkinElmer SCIEX ELAN DRC-e
Forward power (W)	1500
Gas flow rate ($l.min^{-1}$):	
Nebulizer (Ar)	0.8
Auxiliary (Ar)	0.92
Plasma (Ar)	15
Carrier (He)	0.92
Lens voltage	7
Oxide production rate	ThO+/Th+ <0.5 %
Laser Ablation System	New Wave UP193FX
Wavelenght (nm)	193
Energy density on sample (J/cm^2)	4.6–4.7
Repetition rate (Hz)	4
Spot size (µm)	35
Pulse duration (ns)	4
Data acquisition	
Data acquisition protocol	Time resolved analysis
Scanning mode	Peak hopping, 1 point per
Sweeps	1
Reading/Replicate	860
Replicate	1
Background and signal acquisition	~40 s gas blank and ~50–60 s ablation
Dwell time per isotope	20 ms (^{208}Pb and ^{232}Th); 30 ms (^{206}Pb,^{207}Pb, and ^{238}U)
Isotopes analyzed	^{206}Pb, ^{207}Pb, ^{208}Pb, ^{232}Th, ^{238}U
External standard	X36 columbite (Galliski et al. 2021)
Data reduction software	Ioliten (Paton et al. 2011)

Appendix 2

LA–ICP–MS U–Pb in-situ isotope data for columbite–tantalite from Bratislava, Jezuitské Lesy (BM-22), Moravany nad Váhom, Striebornica Ridge (PI-15) and Liešťany, Bystrý Hill (SU-1) granitic pegmatites.

Sample	Point#	Crystal	U (ppm)	2SE	$^{207}Pb/^{235}U$	2SE	$^{206}Pb/^{238}U$	2SE	Rho	$^{206}Pb/^{238}U$ age (Ma)	2SE	$^{207}Pb/^{235}U$ age (Ma)	2SE
BM-22	22nv15c05	cr1	10	0.17	0.500	0.120	0.0558	0.0042	0.06	348	25	346	74
	22nv15c06	cr1	23	0.31	0.494	0.055	0.0566	0.0018	0.14	354	11	367	38
	22nv15c07	cr1	24	0.32	0.447	0.069	0.0571	0.0020	0.06	358	12	353	49
	22nv15c08	cr1	13	0.20	0.264	0.076	0.0578	0.0029	0.10	361	17	190	53
	22nv15c09	cr1	15	0.22	0.930	0.140	0.0568	0.0033	0.35	355	20	587	79
	22nv15c10	cr1	20	0.40	0.557	0.072	0.0568	0.0021	0.18	355	13	389	46
	22nv15c11	cr1	28	0.25	0.458	0.055	0.0559	0.0018	0.16	351	11	347	39
	22nv15c12	cr1	23	0.23	0.481	0.053	0.0558	0.0017	0.10	350	10	352	36
	22nv15c14	cr2	50	0.58	0.790	0.100	0.1078	0.0037	0.04	659	21	564	61
	22nv15c15	cr2	14	0.21	0.710	0.150	0.0571	0.0036	0.04	359	22	456	85
	22nv15c16	cr2	12	0.26	0.510	0.130	0.0539	0.0039	0.06	342	25	346	81
	22nv15c17	cr2	11	0.20	0.480	0.120	0.0580	0.0046	0.11	361	28	327	74
	22nv15c19	cr2	20	0.32	0.500	0.120	0.0550	0.0033	0.02	345	20	343	77
	22nv15c20	cr2	14	0.21	1.610	0.270	0.0604	0.0046	0.37	376	28	860	120
	22nv15c21	cr2	16	0.24	0.600	0.130	0.0552	0.0034	0.05	345	21	394	78
	22nv15c22	cr2	16	0.26	0.950	0.150	0.0550	0.0035	0.02	346	22	604	82
PI-15	22nv15b20	cr1	13	0.21	0.490	0.100	0.0580	0.0032	0.04	362	20	316	61
	22nv15b21	cr1	15	0.45	0.438	0.070	0.0578	0.0024	0.04	361	15	319	48
	22nv15b22	cr1	18	0.9	0.440	0.110	0.0580	0.0044	0.26	362	27	339	83
	22nv15b17	cr2	22	0.39	0.602	0.077	0.0652	0.0023	0.05	407	14	438	51
	22nv15b18	cr2	141	4.70	0.855	0.041	0.0202	0.0006	0.20	129	4	634	22
	22nv15b19	cr2	18	0.20	0.533	0.070	0.0574	0.0020	0.00	359	12	355	44
	22nv15b11	cr3	162	2.00	3.236	0.070	0.0835	0.0011	0.55	517	6	1470	17
	22nv15b12	cr3	123	1.60	1.019	0.042	0.0632	0.0010	0.37	395	6	719	21
	22nv15b13	cr3	25	0.36	0.465	0.097	0.0570	0.0029	0.14	357	18	356	68
	22nv15b08	cr4	32	0.36	0.424	0.058	0.0559	0.0023	0.13	350	14	325	41
	22nv15b09	cr4	17	0.32	0.507	0.075	0.0569	0.0023	0.07	356	14	354	48
	22nv15b10	cr4	34	0.50	0.417	0.053	0.0574	0.0020	0.14	361	12	336	39
	22nv15b05	cr5	23	0.36	7.670	0.680	0.1900	0.0110	0.80	1124	60	2141	84
	22nv15b06	cr5	19	0.38	2.010	0.180	0.0819	0.0033	0.13	507	20	1121	61
	22nv15b07	cr5	23	0.47	0.490	0.120	0.0560	0.0034	0.18	350	20	360	83
SU-1	22nv21b05	cr1	15	0.30	0.870	0.180	0.0543	0.0044	0.11	339	27	550	100
	22nv21b06	cr1	11	0.16	0.553	0.083	0.0556	0.0022	0.14	349	13	334	47
	22nv21b07	cr1	14	0.29	0.500	0.130	0.0532	0.0040	0.18	336	24	337	81
	22nv21b08	cr1	16	0.25	0.341	0.060	0.0467	0.0021	0.14	294	13	269	44
	22nv21b09	cr1	14	0.20	0.540	0.120	0.0568	0.0032	0.17	355	20	364	76
	22nv21b10	cr1	11	0.22	0.660	0.170	0.0465	0.0040	0.26	292	25	418	98
	22nv21b11	cr1	10	0.19	0.560	0.150	0.0550	0.0046	0.05	344	28	350	91
	22nv21b12	cr1	12	0.19	0.510	0.100	0.0566	0.0033	0.01	354	20	354	67

2SE = 2 sigma error; Rho = error correlation of $^{206}Pb/^{238}U$–$^{207}Pb/^{235}U$