

# Monazite-group minerals and xenotime-(Y) in A-type granitic rocks: chemical composition and in-situ Th–U–total Pb EPMA dating (Velence Hills, Hungary)

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## AGEOS

**Abstract:** Accessory monazite-(Ce), -(La), -(Nd), cheralite (monazite-group minerals, MGM) and xenotime-(Y) are rare magmatic accessory minerals in subsolvus A-type granites and related aplite and pegmatite dykes in the Velence Hills, Transdanubian Unit, Hungary. The MGM and xenotime-(Y) form  $\leq 200 \mu\text{m}$  euhedral to subhedral crystals in association with allanite-(Ce), zircon, fluorapatite, and ilmenite. Investigated MGM shows dominant cheralite ( $\text{Ca}^{2+} + \text{Th}^{4+} \leftrightarrow 2\text{REE}^{3+}$ ) and subordinate huttonite ( $\text{REE}^{3+} + \text{P}^{5+} \leftrightarrow \text{Th}^{4+} + \text{Si}^{4+}$ ) substitution with greater extent in pegmatite and aplite than in the host granite. Monazite in-situ Th–U–total Pb electron-microprobe dating of the Velence granite determined  $289 \pm 3 \text{ Ma}$  age (MSWD = 3.22,  $n = 62$ ) and this confirmed post-Variscan, Permian, (Cisuralian) magmatic crystallization. In addition, xenotime-(Y) from the same rock samples gave  $266 \pm 5.2 \text{ Ma}$  age (MSWD = 1.4,  $n = 44$ ), and this corresponds to Permian (Guadalupian) post-magmatic (subsolvus) recrystallization of xenotime-(Y) during a younger event and subsequent overprint of the parental granitic rocks. Although monazite-(Ce) remains relatively unaffected, fluid-induced breakdown of xenotime-(Y) produced numerous tiny thorite and cheralite inclusions in the altered xenotime-(Y) domains which can be formed by coupled dissolution-precipitation reactions between orthomagmatic xenotime-(Y) and younger late to post-magmatic granitic fluids.

**Key words:** monazite, xenotime, EPMA dating, A-type granite, Velence, Hungary

## 1. INTRODUCTION

Monazite-(Ce) and xenotime-(Y) are accessory phosphate minerals and principal carriers of REE and Y in common magmatic and metamorphic rocks, together with allanite-(Ce), zircon, titanite and fluorapatite. Detailed study of their chemical composition and breakdown processes enables understanding of the processes occurring during petrogenesis and evolution of the parental rock (e.g., Montel, 1993; Uher & Broska, 1995; Bea, 1996; Bingen et al., 1996; Montel et al., 1996; Finger et al., 1998; Broska & Siman, 1998; Förster, 1998; Zhu & O'Nions, 1999; Spear & Pyle, 2002; Johan & Johan, 2005; Finger & Krenn, 2007; Krenn & Finger, 2007; Petřík & Konečný, 2009; Ondrejka et al., 2007, 2012; Uher et al., 2015 and many others).

In addition, REE phosphates (monazite and xenotime) are popular targets for petrochronology (Engi, 2017). The commonly high concentrations of Th and U, low initial Pb (Parrish, 1990), low cation diffusion rates (e.g., Cherniak et al., 2004; Cherniak, 2006) allow chemical U–Th–Pb dating by EPMA (e.g., Suzuki & Adachi, 1991; Suzuki et al., 1991; Montel et al., 1996; Scherrer et al., 2000; Cocherie & Albarede, 2001; Williams et al., 2006; Hetherington et al., 2008; Spear et al., 2009). Similarly, in-situ isotopic dating by LA-ICP-MS and SIMS are suited to link ages with their geochemical and textural content (e.g., Kylander-Clark, 2017; Schmitt & Vazquez, 2017).

Monazite-(Ce) and xenotime-(Y) can also be unstable during post-magmatic and metamorphic fluid-activated overprinting

(Budzyń et al., 2010, 2011, 2017, 2018, Budzyń & Sláma, 2019) and both can form subsolvus retrograde allanite-apatite coronas during pervasive fluid-influx (e.g., Broska & Siman, 1998; Finger et al., 1998; Broska et al., 2005; Krenn et al., 2012; Ondrejka et al., 2012, 2016; Upadhyay & Pruseth, 2012; Lo Pò et al., 2016). The redundant amount of Th in monazites in fluid-rock contact can be released and precipitate a further phase (huttonite/thorite, thorianite or cheralite) near the monazite (Harlov et al., 2007; Petřík & Konečný, 2009; Budzyń et al., 2011, 2017; Harlov, 2011; Ondrejka et al., 2012).

However, our knowledge of primary monazite-(Ce) and xenotime-(Y) distribution, their textural and paragenetic relationships and compositional variations is mainly based on the study of these accessory minerals in orogen-related, calc-alkaline S- and I-type granitic suites as well as in orthogneisses and metapelitic to metapsammitic rocks. This also applies to their use as a petrochronological tool for reconstructing their processes and age constraints (e.g., Bea, 1996; Broska et al., 2005, 2012<sup>a</sup>, 2012<sup>b</sup>).

Primary magmatic monazite-(Ce) is a characteristic accessory mineral, especially for peraluminous S-type granites, and it is relatively scarce in subaluminous A- and I-type granitic suites where allanite-(Ce) is a dominant REE phase (Bea, 1996; Broska et al., 2012<sup>a</sup>). In contrast, xenotime-(Y) dominantly occurs in peraluminous leucogranites and granitic pegmatites with S- and A-type affinity and more rarely in I-type granites (Demartin et al., 1991<sup>a</sup>; Buck et al., 1999; Broska et al., 2005;

Hetherington & Harlov, 2008). This is most likely due to the relatively low LREE/(Y + HREE) ratio or Y + HREE enrichment in those suites. Secondary post-magmatic xenotime-(Y) can form during the late-stage activity of hydrothermal fluids in granites or pegmatites during cooling of the host rock (Åmli, 1975; Hetherington & Harlov, 2008; Harlov, 2011).

This paper documents the magmatic and post-magmatic evolution of monazite-group minerals (MGM) and xenotime-(Y) in A-type granites and their aplite and pegmatite derivatives in the Velence granite massif (Hungary). We identify their textural and paragenetic relationships, the variations in chemical compositions and chemical in-situ Th–U–total Pb dating of both minerals. This provides key information on magma crystallization and post-magmatic processes, including rock-fluid interaction and dating of these geological processes, and it enables better understanding of A-type granite evolution.

## 2. GEOLOGICAL SETTING

The Velence granite massif (VGM) is located in the Velence Hills, a small and low mountain in the NW part of Hungary, N of Velence Lake and approximately 70 km SW from Budapest. The Velence Hills rocks belong to the Transdanubian Unit, Inner Western Carpathians (*sensu* Plašienka et al., 1997) where the Balaton Lineament forms the southern tectonic border of the hills. Outcrops of the VGM have eastern and western main areas (Fig. 1).

The VGM intruded into Palaeozoic (Ordovician to Devonian) metamorphic rocks (metasediments) of the Lovas Slate

Formation. The granitoid rocks are divided into two petrographic suites: coarse-grained porphyric monzogranite and fine-grained biotite-bearing monzogranite. The modal composition of both suites is approximately 32 % quartz, 31 % perthitic orthoclase, 32 % plagioclase and 5 % biotite. However, biotite is absent in leucocratic types and the rock colour is light to medium grey (Horváth et al., 2004). The common accessory minerals are fluorapatite, zircon, magnetite, epidote, fluorite, titanite, pyrite and molybdenite (Jantsky, 1957; Buda, 1969, 1985; Buda & Nagy, 1995). The mainly biotite granitic rocks, which commonly have porphyric K-feldspar phenocrysts, are cut by dykes of granite porphyries, aplites and pegmatites (Buda, 1993; Uher & Broska, 1995) and also alkali-ultramafic intrusive bodies including carbonatites and lamprophyres (Horváth et al., 1983). In addition, the VGM in the W and NE part of the Velence Hills is also cut by Eocene volcanites of the Nadap Andesite Formation. The Velence granites have geochemical and tectonic affinity to a post-orogenic or anorogenic A-type granite suite (Uher & Broska, 1996; Broska & Uher, 2001; Uher et al., 2002), and U–Pb zircon dating constrained their Permian age to  $\approx 280$  Ma (Uher et al., 2009). Various Permian magmatic intrusions occurred in the Inner Western Carpathians: intermediate and basic volcanism (Vozárová et al., 2018), lamprophyre (Spišiak et al., 2018) and granite (Broska & Kubiš, 2018).

## 3. ANALYTICAL METHODS AND SAMPLE LOCATIONS

The MGM and xenotime-(Y) were analysed by CAMECA SX-100 microprobe at the Department of Electron Microanalysis

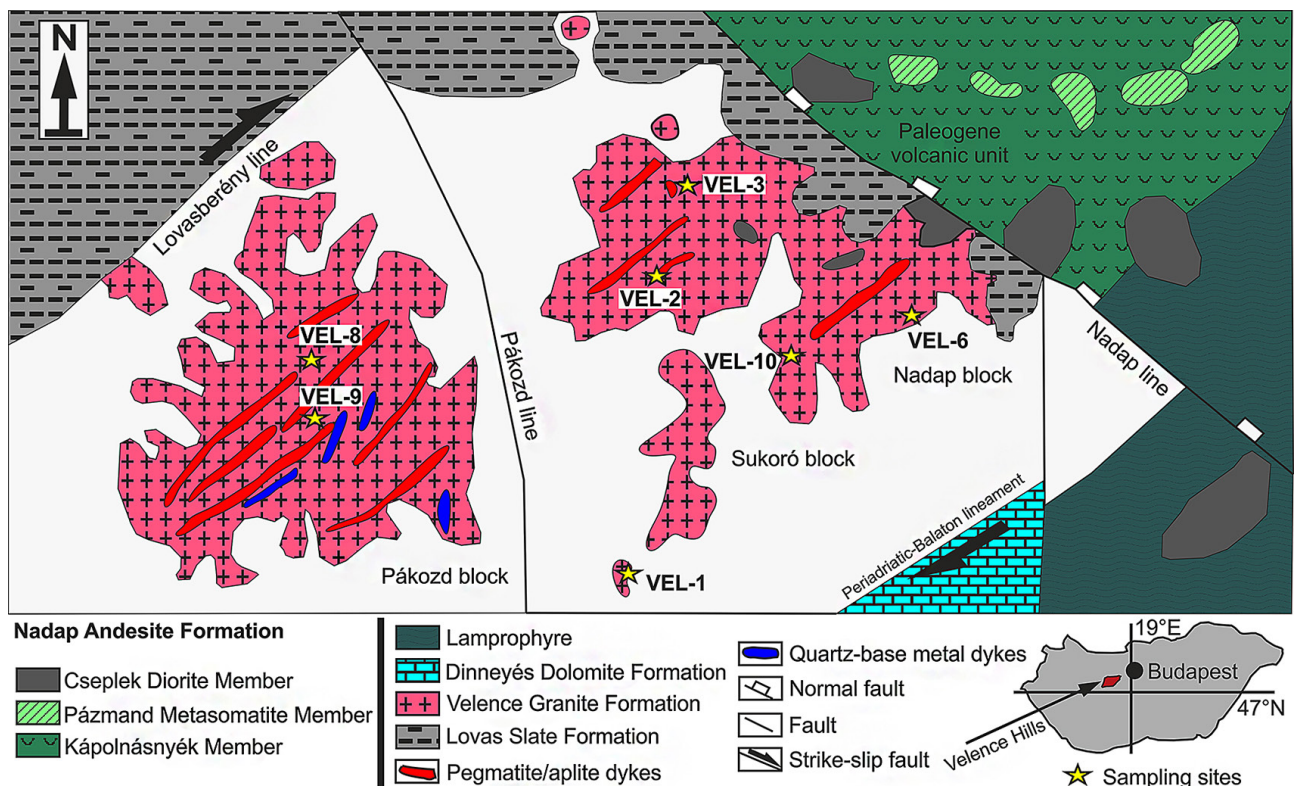


Fig. 1: Geological map of the Velence Hills area with sample locations (modified after Horváth et al., 2004 and Benkó et al., 2010, 2014<sup>a</sup>, 2014<sup>b</sup>).

**Table 1:** Localisation of samples and GPS coordinates.

Sample	Localisation and description of area	Rock type	GPS coordinates
VEL-1	Natural outcrop southwest from Sukoró village	porphyritic granite	N: 47°12.636' E: 18°34.582'
VEL-2F	Outcrop next to military monument near Sukoró village	porphyritic granite	N: 47°13.068' E: 18°34.851'
VEL-2G	Outcrop next to military monument near Sukoró village	banded aplite	N: 47°12.636' E: 18°34.851'
VEL-3B	Road outcrop next to Sukoró village	aplite	N: 47°13.216' E: 18°34.790'
VEL-6C	Outcrop behind the transmitter near the road on the E slope of Bence Hill near Nadap village	microgranite	N: 47°19.942' E: 18°37.615'
VEL-8A	Béllá valley near Pákozsd village	aplite	N: 47°12.636' E: 18°31.38'
VEL-8B	Béllá valley near Pákozsd village	aplite	N: 47°12.636' E: 18°31.38'
VEL-8C	Béllá valley near Pákozsd village	aplite	N: 47°12.636' E: 18°31.38'
VEL-9	Abandoned fluorite quarry near Pákozsd village	pegmatite	N: 47°13.721' E: 18°31.425'
VEL-9A	Abandoned fluorite quarry near Pákozsd village	pegmatite	N: 47°13.721' E: 18°31.425'
VEL-10	Abandoned Rigohégy quarry near Sukoró village	biotite granite	N: 47°14.451' E: 18°34.926'

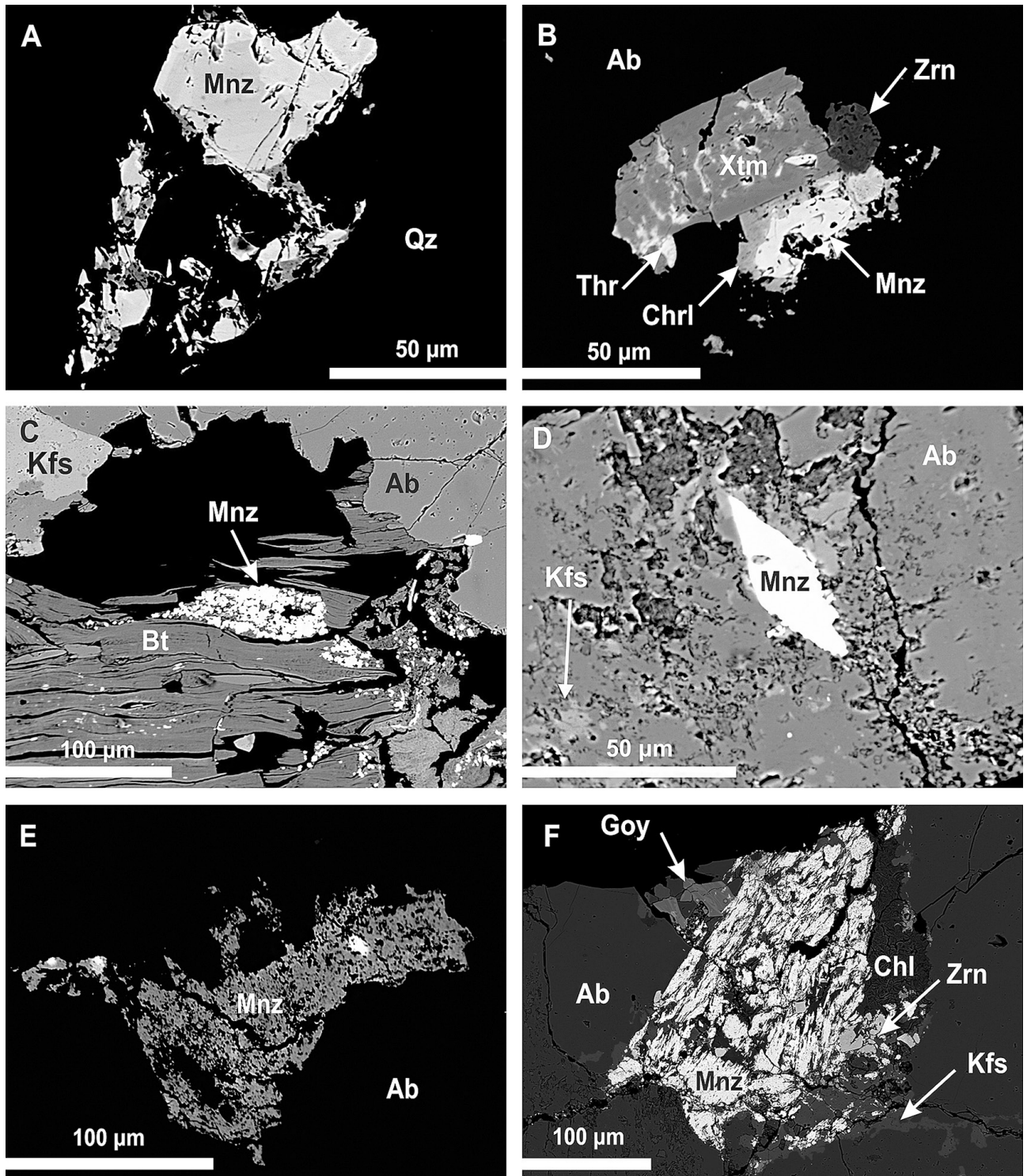
at the State Geological Institute of Dionýz Štúr in Bratislava. The analytical conditions used for non-dated mineral phases included 15 kV accelerating voltage, 35–40 nA probe current, 3–10  $\mu\text{m}$  beam diameter and dependent on mineral sensitivity to induced beam damage.

Monazite and xenotime Th–U–total Pb dating conditions comprised 15 kV accelerating voltage, 180 nA probe current and 3  $\mu\text{m}$  beam diameter. Finally, the PAP matrix correction factors were used, and the following conditions are specified in (Tab. 2.) Analytical lines free of overlap were preferentially selected. Mutual interferences among REE were corrected by empirical correction factors. The PbM $\alpha$  line is overlapped by LaL $\alpha$ , ThM $\zeta$ 1, ThM $\zeta$ 2, and YL $\gamma$ 2,3. The weak interference with LaL $\alpha$  was neglected. The UM $\beta$  line is overlapped by ThM $\zeta$ , ThM3-N4 and ThM5-P3. The PbM $\alpha$  line overlaps were resolved with consideration of curved exponential background. Monazite dating includes measurement of 7–9 age monazite standards and applying of correction dependencies derived from measured compositions. Further details and description of dating procedure named MARC can be found in Konečný et al. (2018). Procedure of xenotime-(Y) dating was slightly modified because of using the PbM $\beta$ . Back-scattered-electron imaging (BSE) was performed by CAMECA SX-100 microprobe at the State Geological Institute of Dionýz Štúr in Bratislava and the JEOL JXA-8530F microprobe at the Earth Science Institute of the Slovak Academy of Sciences in Banská Bystrica. The formula element contents are expressed in atoms per formula unit (apfu), and the MGM and xenotime-(Y) formulae were normalised on 4 oxygen anions.

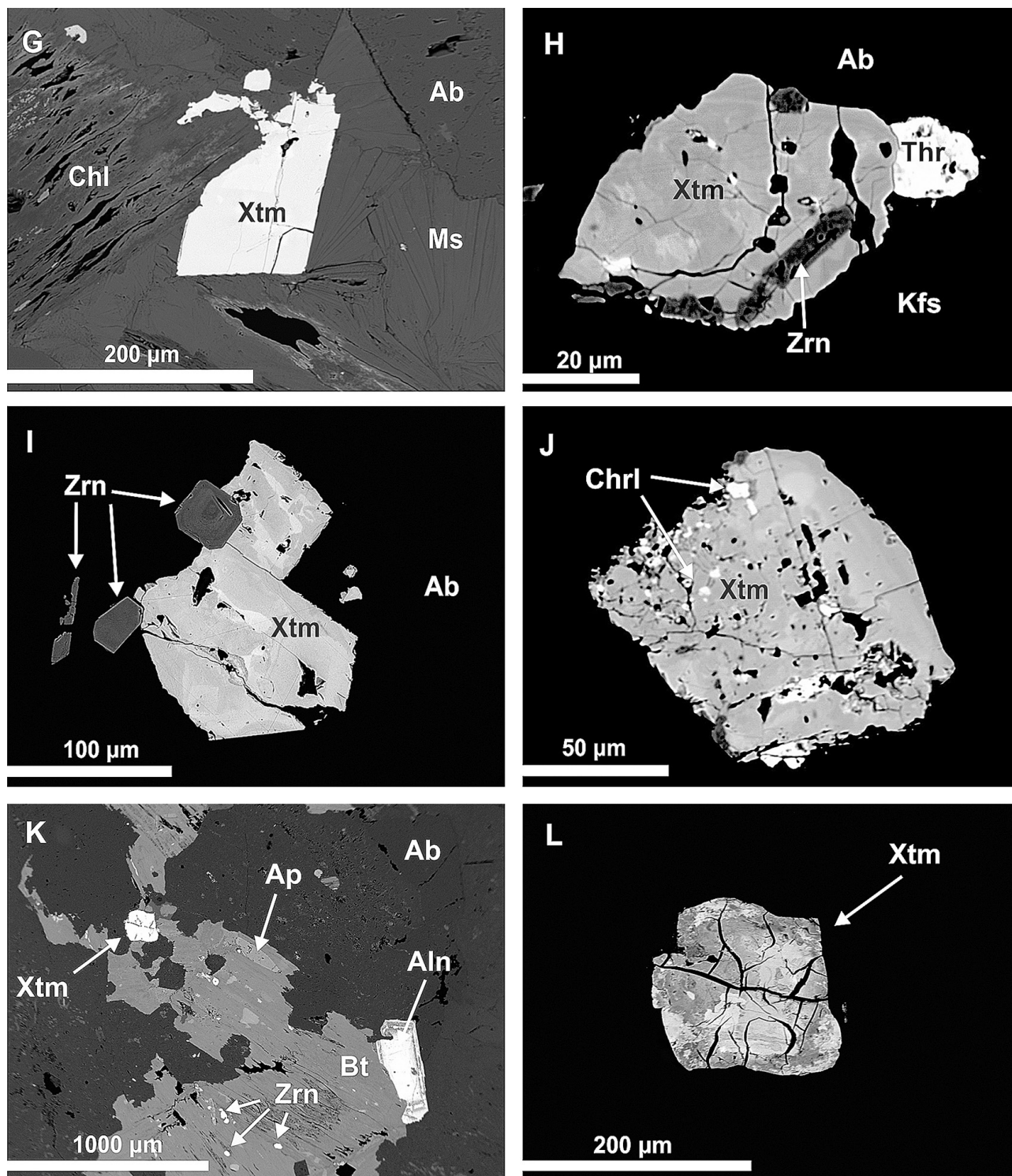
The investigated samples were collected from 7 different localities. The VEL-1, 2, 3, 6, and 10 samples came from the eastern granite body in the Sukoró area and the VEL-8 and 9 samples from the western granite body in the Pákozsd area. Granite samples include leucocratic coarse-grained porphyritic granite with K-feldspar, albite, quartz, and/or biotite. The aplite samples comprise fine-grained quartz, feldspar, and/or biotite, and only one sample contains granitic pegmatite with unequigranular texture and K-feldspar porphyritic phenocrysts. The location of sampling sites with GPS coordinates is presented in Tab. 1.

**Table 2:** EPMA conditions used for each element.

Element	Line	Detection limit (3 $\sigma$ ) in ppm			MGM and xenotime-(Y)
		Crystal	Calibrant		
S	K $\alpha$	LPET	baryte		81-262
P	K $\alpha$	LPET	apatite		105-322
As	L $\alpha$	TAP	GaAs		485-677
Si	K $\alpha$	TAP	wollastonite		127-363
Th	M $\alpha$	LPET	ThO <sub>2</sub>		218-569
U	M $\beta$	LPET	UO <sub>2</sub>		195-835
Al	K $\alpha$	TAP	Al <sub>2</sub> O <sub>3</sub>		127-379
Y	L $\alpha$	LPET	YPO <sub>4</sub>		161-543
La	L $\alpha$	LLIF	LaPO <sub>4</sub>		644-1295
Ce	L $\alpha$	LLIF	CePO <sub>4</sub>		572-1147
Pr	L $\beta$	LLIF	PrPO <sub>4</sub>		977-1178
Nd	L $\alpha$	LLIF	NdPO <sub>4</sub>		615-1003
Sm	L $\alpha$	LLIF	SmPO <sub>4</sub>		722-1247
Eu	L $\beta$	LLIF	EuPO <sub>4</sub>		1162-1410
Gd	L $\alpha$	LLIF	GdPO <sub>4</sub>		759-1155
Tb	L $\alpha$	LLIF	TbPO <sub>4</sub>		687-1235
Dy	L $\beta$	LLIF	DyPO <sub>4</sub>		1417-1741
Ho	L $\beta$	LLIF	HoPO <sub>4</sub>		1537-1850
Er	L $\beta$	LLIF	ErPO <sub>4</sub>		1629-1978
Tm	L $\alpha$	LLIF	TmPO <sub>4</sub>		788-1393
Yb	L $\alpha$	LLIF	YbPO <sub>4</sub>		851-1387
Lu	L $\beta$	LLIF	LuPO <sub>4</sub>		1541-3066
Ca	K $\alpha$	LPET	wollastonite		79-263
Sr	L $\alpha$	LPET	SrTiO <sub>3</sub>		272-490
Ba	L $\alpha$	LPET	baryte		417-480
Fe	K $\alpha$	LIF	fayalite		402-1159
Mn	K $\alpha$	LLIF	rhodonite		902-1086
Pb	M $\alpha$ , $\beta$	LPET	PbCO <sub>3</sub>		92-677
Na	K $\alpha$	TAP	albite		410-683
K	K $\alpha$	LPET	orthoclase		85-251
F	K $\alpha$	LPCO	CaF <sub>2</sub>		584-754
Cl	K $\alpha$	LPET	NaCl		232-271



**Fig. 2:** Back-scattered electron (BSE) images of MGM and associated minerals. A – Anhedral aggregate of monazite-(Ce) – (Mnz) in quartz (Qz), where brighter domains are Th-rich. Sample VEL-2F (granite). B – Accessory mineral assemblage of monazite-(Ce) – (Mnz), xenotime-(Y) – (Xtm) with cheralite (Chrl) and thorite (Thr) inclusions and zircon (Zrn) in albite (Ab). Sample VEL-2F (granite). C – Rare glomeroporphyric aggregate (synneusis) of monazite-(Ce) – (Mnz) in biotite (Bt). Sample VEL-2F (granite). D – Solitary euhedral crystal of monazite-(Ce) – (Mnz) in albite (Ab). Sample VEL-2F (granite). E – Anhedral aggregate of MGM (Mnz) in albite (Ab). Sample VEL-9 (pegmatite). F – Large aggregate of hydrated(?) MGM (Mnz) or possible rhabdophane in association with zircon (Zrn), albite (Ab) and K-feldspar (Kfs). Secondary minerals are represented by chlorite (Chl) and rare secondary goyazite (Goy). Sample VEL-9 (pegmatite). Mineral abbreviations are after Whitney & Evans (2010).



**Fig. 2:** continued – Back-scattered electron (BSE) images of xenotime-(Y) and associated minerals. G – Subhedral xenotime-(Y) – (Xtm) in association with partly chloritised (Chl) muscovite (Ms) and albite (Ab). Sample VEL-1 (granite). H – Heterogeneous crystal of xenotime-(Y) – (Xtm) with zircon inclusions (Zrn) and thorite (Thr). Rock-forming minerals are represented by albite (Ab) and K-feldspar (Kfs). Sample VEL-2F (granite). I – Typical isostructural relationship of accessory xenotime-(Y) – (Xtm) and zircon (Zrn) in albite (Ab). Xenotime-(Y) shows brighter Th, U-rich convolute zonation. Sample VEL-1 (granite). J – Large xenotime-(Y) crystal (Xtm) with secondary irregular zonation and porosity and numerous inclusions of cheralite (Chrl). Sample VEL-2F (granite). K – Accessory mineral assemblage of xenotime-(Y) – (Xtm), allanite-(Ce) – (Aln), fluorapatite (Ap) and zircon (Zrn) scattered in biotite (Bt) and albite (Ab). Sample VEL-9 (pegmatite). L – Detail of image K. Heterogeneous composition and secondary irregular zonation of xenotime-(Y) – (Xtm) with developed system of cracks and fissures. Sample VEL-9 (pegmatite). Mineral abbreviations are after Whitney & Evans (2010).

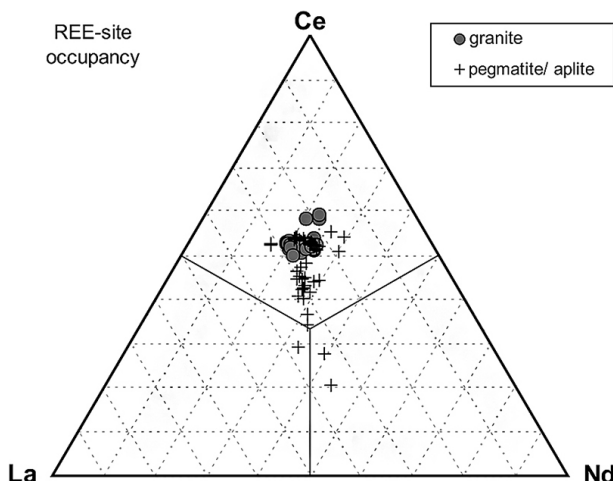


Fig. 3: Ternary diagram Ce–La–Nd showing MGM endmembers in parental granite and pegmatite/aplite with contrasting Ce, La and Nd distribution (atomic proportions).

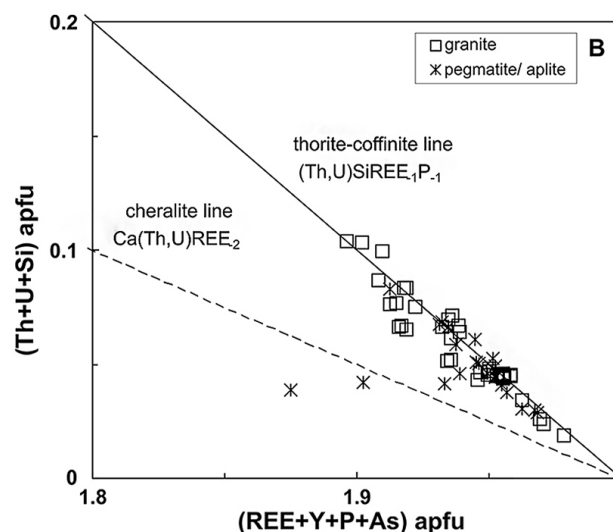
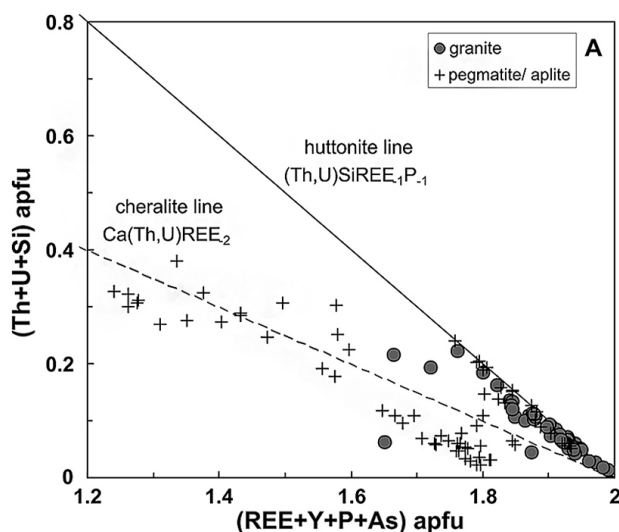


Fig. 4: Composition of accessory minerals in Th + U + Si vs. REE + Y + P + As substitution diagram (atomic proportions) with ideal cheralite and huttonite or thorite-coffinite substitution vectors (straight lines). A – MGM, B – xenotime-(Y).

## 4. RESULTS AND MINERAL COMPOSITION

### 4.1. Monazite-group minerals

The MGM form euhedral to anhedral solitary crystals and occasional aggregates, and their size mostly ranges from 20 to 200  $\mu\text{m}$  (Fig. 2). The MGM has mostly oscillatory or irregular zoning visible on back-scattered electron (BSE) images and is generally associated with accessory xenotime-(Y) and zircon (Fig. 2B). It occasionally forms glomeroporphyric clusters (synnesis) scattered in the biotite and interstitial spaces (Fig. 2C) or euhedral crystals enclosed in feldspars (Fig. 2D). However, large fragmented MGM aggregates in close association with goyazite  $\text{SrAl}_3(\text{PO}_4)_2(\text{OH})_5 \cdot (\text{H}_2\text{O})$ , zircon and chlorite in feldspars are rarely present (Fig. 2F).

The investigated MGM are mostly Ce-dominant (up to 32.3 wt.%  $\text{Ce}_2\text{O}_3$ ; 0.47 apfu Ce; Tab. 1). However, minor cheralitic monazite-(Nd) with Ce/Nd = 0.47 and monazite-(La) with

Ce/La = 0.78 also occur in the pegmatite (Fig. 3, Tab. 3). Yttrium concentration is relatively high (up to 12.7 wt.%  $\text{Y}_2\text{O}_3$ ; 0.27 apfu Y) and there are also minor concentrations of heavy REE (HREE = Gd to Lu), with Gd the most significant (up to 4.9 wt.%  $\text{Gd}_2\text{O}_3$ ; 0.06 apfu Gd).

Thorium and Ca contents vary greatly in MGM (0.53 to 34.9 wt.%  $\text{ThO}_2$ ; up to 0.31 apfu Th; 0.1 to 10.7 wt.% CaO and 0.51 apfu Ca). These concentrations increase in MGM originating from pegmatite compared to that from granite, and MGM from pegmatite/aplite is generally richer in cheralite ( $\text{Ca}_{0.5}\text{Th}_{0.5}\text{PO}_4$ ) and huttonite ( $\text{ThSiO}_4$ ) components. Cheralite endmember occasionally occurs (Fig. 4A, 5), but is restricted to pegmatite/aplite only. The MGM from pegmatite has slightly increased F content up to 1.1 wt.% F, but the low totals suggest that the phases are most likely hydrated.

The MGM mole fractions are:  $X_{\text{mnz}} = 71 - 98$ ;  $X_{\text{hutt}} = 0 - 17$ ;  $X_{\text{chrl}} = 0 - 68$  (Fig. 5). There is also depleted light REE (LREE

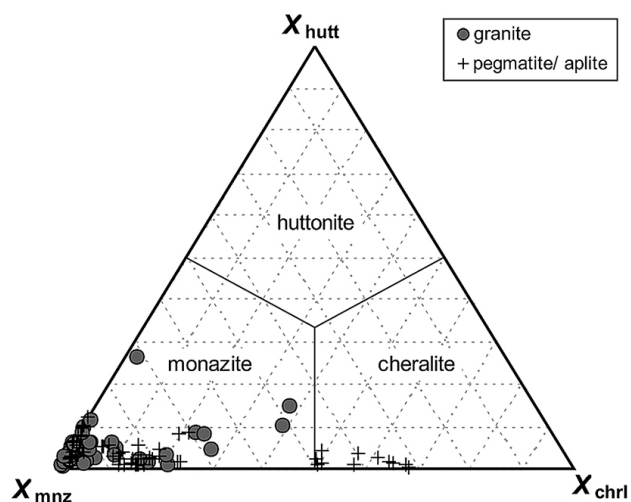


Fig. 5: Ternary diagram of MGM mole fractions ( $X_{\text{mnz}}$  = monazite,  $X_{\text{hutt}}$  = huttonite,  $X_{\text{chrl}}$  = cheralite) in parental granite and pegmatite/aplite with contrasting mole fractions distribution.

= La to Sm) content at the expense of other minor elements, mostly Th and Ca. The Ce deficiency relative to other LREE in the pegmatite/aplite is evident in the REE mineral/chondrite

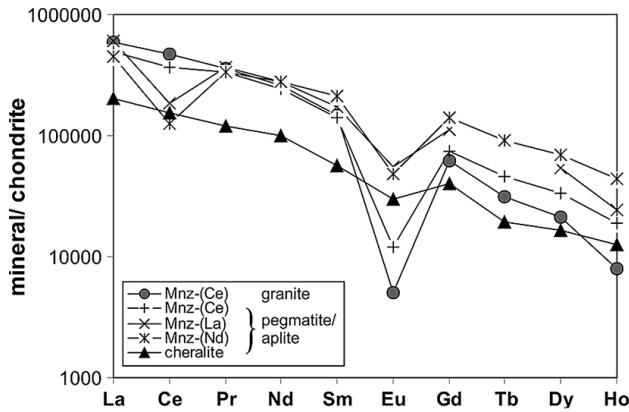


Fig. 6: Chondrite-normalised patterns of selected REE in MGM from parental granite and pegmatite/aplite. Normalised values after Barrat et al. (2012).

normalised patterns (Fig. 6). The compositional X-ray mapping shows that monazite-(Ce) crystal from the host granite has relatively homogeneous REE distribution and Th enrichment in marginal areas (Fig. 7). Other trace elements (As, S, Sr, Ba and Cl) are generally very low or below EPMA detection limit (Tab. 3).

#### 4.2. Xenotime-(Y)

Accessory xenotime-(Y) is the main carrier of HREE in the VGM but it has relatively less abundance than MGM and occurs as anhedral to subhedral crystals up to 200 µm in size. Xenotime-(Y) is associated with zircon, cheralite, allanite-(Ce) and fluorapatite (Fig. 2), and most crystals have numerous tiny inclusions of Th-silicate (thorite?) or Ca-Th-phosphate (cheralite) and show irregular zoning reflecting the variable REE/Th-Ca ratio (Fig. 2H,J). There are also irregularly shaped areas within the crystals and forming aureoles around the Th-Ca inclusions. The darker areas are richer in REE and these record relative depletion of Th and partly also U. Finally, the secondary zone development

Table 3: Representative compositions of EPMA of MGM (wt. %).

Sample	VEL-1	VEL-2F	VEL-2G	VEL-3B	VEL-8A	VEL-8C	VEL-9	VEL-9A	VEL-9A	VEL-9A
End member	Mnz-(Ce)	Mnz-(Ce)	Mnz-(Ce)	Mnz-(Ce)	Mnz-(Ce)	Mnz-(Ce)	Mnz-(Nd)	Mnz-(La)	Chrl	Chrl
SO <sub>3</sub>	0.02	0.01	0.02	0.03	0.02	0.02	0.24	0.18	0.08	0.21
P <sub>2</sub> O <sub>5</sub>	28.94	26.09	26.95	25.11	27.80	27.43	28.09	28.98	27.75	28.56
As <sub>2</sub> O <sub>5</sub>	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.15	0.20	0.00
SiO <sub>2</sub>	0.61	2.88	2.51	2.99	1.46	2.55	0.48	1.54	2.18	0.67
ThO <sub>2</sub>	4.41	11.21	9.86	12.28	9.65	5.74	11.07	4.52	18.57	28.91
UO <sub>2</sub>	0.05	0.44	0.40	0.29	0.17	0.75	0.21	0.00	0.18	0.20
Al <sub>2</sub> O <sub>3</sub>	0.00	0.00	0.14	0.00	0.00	0.00	0.35	1.05	0.00	0.00
Y <sub>2</sub> O <sub>3</sub>	2.03	3.79	3.04	0.90	0.70	3.11	5.26	4.57	3.43	2.78
La <sub>2</sub> O <sub>3</sub>	13.74	10.34	11.57	12.81	16.90	7.90	10.63	14.14	8.66	7.06
Ce <sub>2</sub> O <sub>3</sub>	28.75	22.81	25.91	27.52	28.80	26.26	8.96	11.03	18.01	12.81
Pr <sub>2</sub> O <sub>3</sub>	3.31	2.88	3.17	3.20	2.89	3.83	3.08	3.37	1.90	1.60
Nd <sub>2</sub> O <sub>3</sub>	12.15	11.50	12.13	11.24	8.96	14.67	12.86	12.87	8.39	5.40
Sm <sub>2</sub> O <sub>3</sub>	2.30	2.63	2.61	1.73	1.11	3.19	3.11	2.66	1.77	0.57
Eu <sub>2</sub> O <sub>3</sub>	0.06	0.00	0.01	0.03	0.06	0.00	0.31	0.32	0.00	0.18
Gd <sub>2</sub> O <sub>3</sub>	1.58	1.80	2.01	0.57	0.46	1.75	2.38	2.32	1.74	1.02
Tb <sub>2</sub> O <sub>3</sub>	0.14	0.23	0.20	0.05	0.07	0.19	0.20	0.00	0.21	0.16
Dy <sub>2</sub> O <sub>3</sub>	0.70	0.94	0.89	0.23	0.18	0.96	1.36	1.36	0.92	0.54
Ho <sub>2</sub> O <sub>3</sub>	0.05	0.15	0.13	0.00	0.00	0.08	0.11	0.14	0.11	0.08
Er <sub>2</sub> O <sub>3</sub>	0.46	0.62	0.51	0.35	0.25	0.34	0.63	0.38	0.38	0.50
Tm <sub>2</sub> O <sub>3</sub>	0.11	0.12	0.11	0.09	0.01	0.06	0.12	0.27	0.00	0.24
Yb <sub>2</sub> O <sub>3</sub>	0.16	0.22	0.15	0.14	0.13	0.20	0.35	0.31	0.00	0.33
Lu <sub>2</sub> O <sub>3</sub>	0.08	0.07	0.13	0.10	0.15	0.08	0.11	0.36	0.00	0.25
CaO	0.47	0.35	0.19	0.09	1.03	0.11	4.20	3.71	4.02	6.82
SrO	0.01	0.00	0.03	0.00	0.01	0.01	0.08	0.00	0.00	0.18
FeO total	0.00	0.02	0.06	0.00	0.82	0.43	2.09	0.00	0.90	0.63
PbO	0.06	0.13	0.13	0.16	0.13	0.10	1.63	1.47	0.08	0.11
F	0.00	0.00	0.00	0.00	0.00	0.00	0.44	1.12	0.00	0.00
Total	100.17	99.24	102.86	99.90	101.76	99.76	98.36	96.82	99.47	99.82

Table 3: continued - Representative compositions of EPMA of MGM (apfu).

Sample	VEL-1	VEL-2F	VEL-2G	VEL-3B	VEL-8A	VEL-8C	VEL-9	VEL-9A	VEL-9A	VEL-9A
End member	Mnz-(Ce)	Mnz-(Ce)	Mnz-(Ce)	Mnz-(Ce)	Mnz-(Ce)	Mnz-(Ce)	Mnz-(Nd)	Mnz-(La)	Chrl	Chrl
S <sup>6+</sup>	0.001	0.000	0.001	0.001	0.001	0.001	0.007	0.005	0.002	0.006
P <sup>5+</sup>	0.968	0.891	0.895	0.873	0.926	0.915	0.915	0.904	0.914	0.943
As <sup>5+</sup>	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.003	0.004	0.000
Si <sup>4+</sup>	0.024	0.116	0.098	0.123	0.058	0.100	0.019	0.057	0.085	0.026
Th <sup>4+</sup>	0.040	0.103	0.088	0.115	0.086	0.051	0.097	0.038	0.165	0.256
U <sup>4+</sup>	0.000	0.004	0.003	0.003	0.001	0.007	0.002	0.000	0.002	0.002
Al <sup>3+</sup>	0.000	0.000	0.006	0.000	0.000	0.000	0.016	0.046	0.000	0.000
Y <sup>3+</sup>	0.043	0.081	0.064	0.020	0.015	0.065	0.108	0.090	0.071	0.058
La <sup>3+</sup>	0.200	0.154	0.167	0.194	0.245	0.115	0.151	0.192	0.124	0.102
Ce <sup>3+</sup>	0.416	0.337	0.372	0.414	0.415	0.379	0.126	0.149	0.257	0.183
Pr <sup>3+</sup>	0.048	0.042	0.045	0.048	0.041	0.055	0.043	0.045	0.027	0.023
Nd <sup>3+</sup>	0.172	0.166	0.170	0.165	0.126	0.207	0.177	0.169	0.117	0.075
Sm <sup>3+</sup>	0.031	0.037	0.035	0.025	0.015	0.043	0.041	0.034	0.024	0.008
Eu <sup>3+</sup>	0.001	0.000	0.000	0.000	0.001	0.000	0.004	0.004	0.000	0.002
Gd <sup>3+</sup>	0.021	0.024	0.026	0.008	0.006	0.023	0.030	0.028	0.022	0.013
Tb <sup>3+</sup>	0.002	0.003	0.003	0.001	0.001	0.002	0.003	0.000	0.003	0.002
Dy <sup>3+</sup>	0.009	0.012	0.011	0.003	0.002	0.012	0.017	0.016	0.011	0.007
Ho <sup>3+</sup>	0.001	0.002	0.002	0.000	0.000	0.001	0.001	0.002	0.001	0.001
Er <sup>3+</sup>	0.006	0.008	0.006	0.005	0.003	0.004	0.008	0.004	0.005	0.006
Tm <sup>3+</sup>	0.001	0.002	0.001	0.001	0.000	0.001	0.001	0.003	0.000	0.003
Yb <sup>3+</sup>	0.002	0.003	0.002	0.002	0.002	0.002	0.004	0.003	0.000	0.004
Lu <sup>3+</sup>	0.001	0.001	0.002	0.001	0.002	0.001	0.001	0.004	0.000	0.003
Ca <sup>2+</sup>	0.020	0.015	0.008	0.004	0.043	0.005	0.173	0.147	0.168	0.285
Si <sup>2+</sup>	0.000	0.000	0.001	0.000	0.000	0.000	0.002	0.000	0.000	0.004
Fe <sup>2+</sup>	0.000	0.001	0.002	0.000	0.027	0.014	0.067	0.000	0.029	0.020
Pb <sup>2+</sup>	0.001	0.001	0.001	0.002	0.001	0.001	0.017	0.015	0.001	0.001
Position X	0.993	1.008	0.994	0.997	0.984	1.016	0.940	0.969	1.005	0.975
Position A	1.013	0.996	1.016	1.008	1.034	0.990	1.089	0.989	1.026	1.058
Total	2.005	2.004	2.010	2.005	2.018	2.006	2.030	1.958	2.032	2.033

indicates xenotime-(Y) alteration and secondary thorite and cheralite formation (Fig. 2G, J).

In addition to Y (up to 46.7 wt.% Y<sub>2</sub>O<sub>3</sub>; 0.80 apfu Y), xenotime-(Y) also has elevated HREE concentrations. These are principally Dy (up to 7.0 wt.% Dy<sub>2</sub>O<sub>3</sub>; 0.08 apfu Dy) and Yb (up to 5.8 wt.% Yb<sub>2</sub>O<sub>3</sub>; 0.06 apfu Yb). The LREE are not readily incorporated into the xenotime-(Y) structure, and consequently have max. value of only 2.65 wt.% LREE<sub>2</sub>O<sub>3</sub> (0.03 apfu LREE).

The actinide elements (Th, U) vary in concentration but Th > U (up to 4.4 wt.% ThO<sub>2</sub>; 0.034 apfu Th and up to 2.0 wt.% UO<sub>2</sub>; 0.02 apfu U; Fig. 8, Tab. 4). There is also positive correlation between Th+U+Si and REE+P (apfu), and this indicates coupled substitution corresponding to the thorite and coffinite (Th+U)SiREE<sub>1</sub>P<sub>1</sub> substitution vector (Fig 4B).

The chemical composition of xenotime-(Y) and normalised REE patterns have no significant difference in parental granite and pegmatite/aplite. Both REE patterns show a weakly-developed M-type tetrad effect in HREE (granite: T3 = 1.22,

T4 = 0.98, combined T3-4 = 1.1; pegmatite/aplite: T3 = 1.27, T4 = 1.04, combined T3-4 = 1.15; calculation according to Irber (1999), see Fig. 9. In addition, X-ray elemental mapping of xenotime-(Y) crystal from the parental granite reveals relatively homogeneous distribution of REE, and Ca, Th and Si distribution suggests the presence of tiny cheralite inclusions (Fig. 10). Other trace elements (As and S) are generally very low or below the EPMA detection limit (Tab. 4).

#### 4.3. Monazite-(Ce) and xenotime-(Y) in-situ chemical dating

Non-stoichiometric and altered Th-Ca-rich MGM have low totals and they were therefore omitted from the age calculation. The applied dating method gave the following ages: 289 ± 2.9 Ma (monazite, n = 62, MSWD: 3.22; Fig. 11, Tab. 5.) and 266 ± 5.2 Ma (xenotime, n = 44, MSWD: 1.4; Fig. 12, Tab. 6.). The data also elucidated systematic primary Permian (Cisuralian) magmatic age for granitic intrusion emplacement



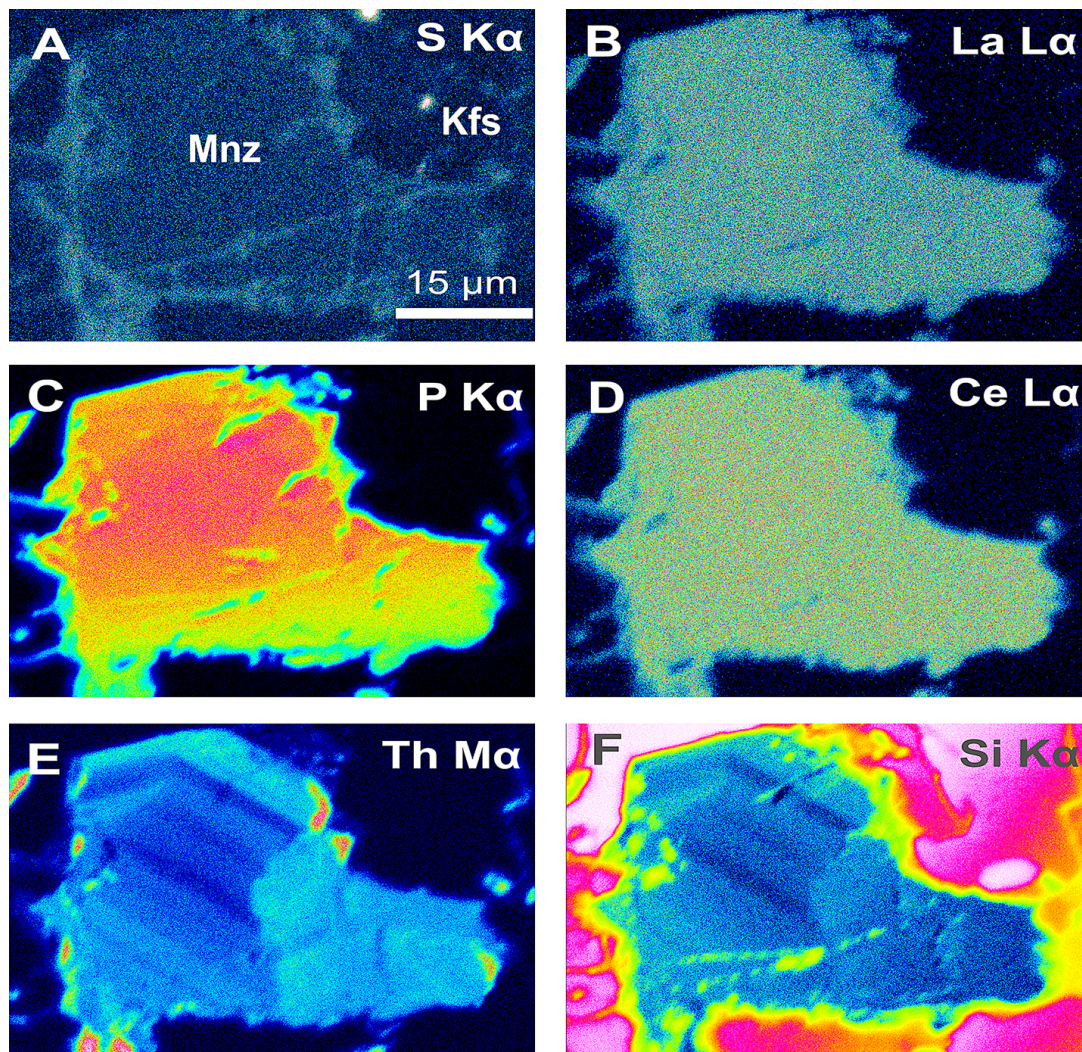


Fig. 7: False colour X-ray mapping showing the distribution of chemical elements in MGM. False colours: blue – very low, green – low, yellow – medium, orange – high, red – very high.

and Permian (Guadalupian) crystallization of the apparently younger xenotime-(Y). Moreover, the sporadic occurrence of 670 – 640 Ma old monazite-(Ce) domains in the Velence

granite was detected in all dated samples (n = 11). However, the detailed comparative study of EPMA in-situ chemical dating spots shows no age differences between MGM and xenotime-(Y) populations from parental granite and those from pegmatite/aplite.

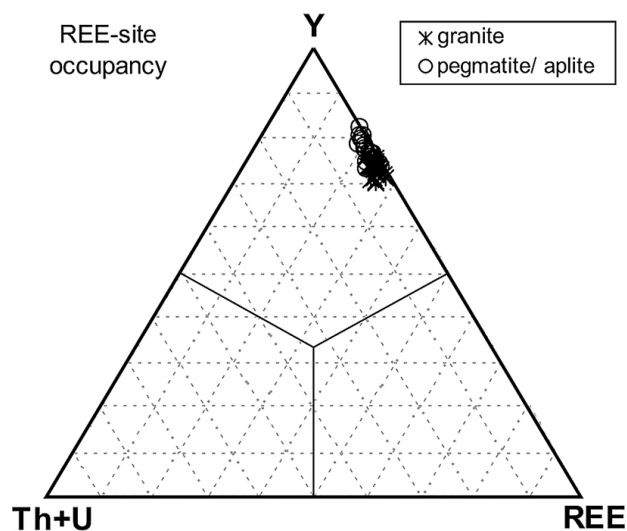


Fig. 8: Ternary diagram Y-REE-(Th+U) showing xenotime-(Y) composition in parental granite and pegmatite/aplite.

## 5. DISCUSSION AND CONCLUSIONS

### 5.1. Monazite-group minerals and xenotime-(Y) composition and typomorphic distribution

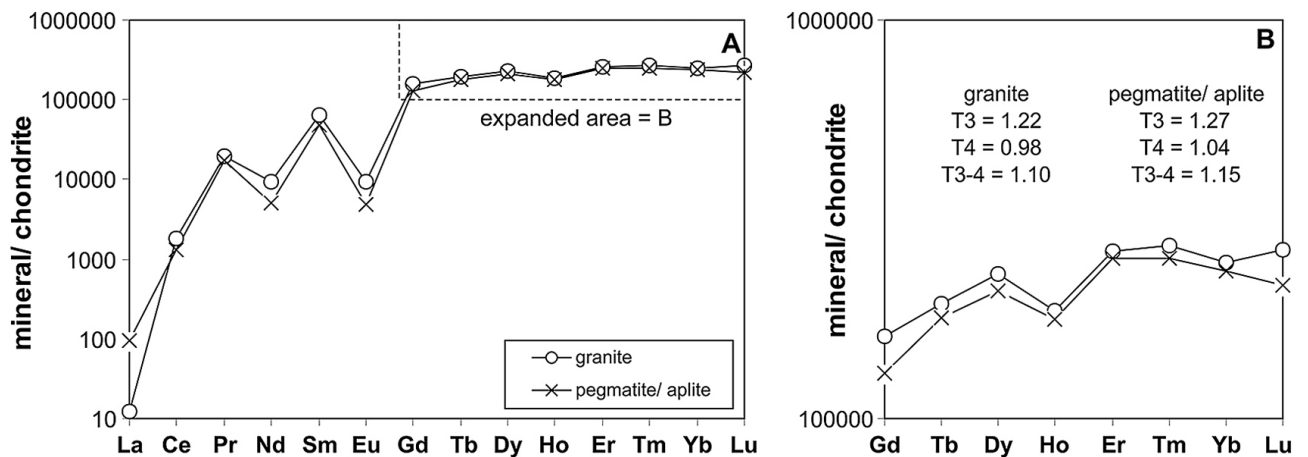
Monazite-(Ce) and xenotime-(Y) are important accessory REE-bearing minerals and reliable petrogenetic indicators of granitic rock evolutionary history. In particular, monazite-(Ce) is the most common accessory LREE-rich mineral phase of peraluminous and low-Ca granitic suites, commonly with S-type, or highly evolved I-type affinity (e.g., Broska & Uher, 1991; Bea, 1996; Förster, 1998; Kelts et al., 2008; Petřík & Konečný, 2009; Broska et al., 2012<sup>b</sup>; Uher et al., 2014). REE phosphates are almost absent in hypersolvus A-type granites, but they sporadically occur in the subsolvus type with allanite-(Ce) the dominant REE-bearing phase (e.g., Uher & Broska,

**Table 4:** Representative compositions of EPMA of xenotime-(Y) (wt.%).

Sample	VEL-1	VEL-1	VEL-1	VEL-2F	VEL-2F	VEL-6C	VEL-8A	VEL-8B	VEL-8B	VEL-8C	VEL-10
End member	Xtm	Xtm	Xtm	Xtm	Xtm	Xtm	Xtm	Xtm	Xtm	Xtm	Xtm
SO <sub>3</sub>	0.01	0.03	0.01	0.02	0.02	0.12	0.00	0.02	0.01	0.01	0.02
P <sub>2</sub> O <sub>5</sub>	33.83	33.24	32.37	34.64	34.63	34.98	32.95	33.72	33.18	34.75	32.61
As <sub>2</sub> O <sub>5</sub>	0.00	0.00	0.00	0.01	0.03	0.10	0.07	0.10	0.03	0.06	0.00
SiO <sub>2</sub>	1.05	1.60	1.47	0.74	0.93	1.21	1.63	1.53	4.15	0.71	1.18
ThO <sub>2</sub>	0.79	2.56	2.39	1.10	0.84	0.16	2.16	1.09	0.19	0.35	0.50
UO <sub>2</sub>	0.84	1.63	1.76	1.23	1.78	0.15	1.49	1.06	0.73	0.49	1.03
Al <sub>2</sub> O <sub>3</sub>	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.79	0.00	0.00
Y <sub>2</sub> O <sub>3</sub>	40.17	38.16	37.65	39.67	39.66	46.62	41.21	43.21	38.81	42.62	39.43
La <sub>2</sub> O <sub>3</sub>	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Ce <sub>2</sub> O <sub>3</sub>	0.06	0.14	0.12	0.08	0.11	0.11	0.09	0.07	0.12	0.10	0.07
Pr <sub>2</sub> O <sub>3</sub>	0.21	0.24	0.17	0.21	0.18	0.14	0.16	0.14	0.13	0.17	0.14
Nd <sub>2</sub> O <sub>3</sub>	0.38	0.69	0.72	0.37	0.46	0.12	0.25	0.34	0.54	0.20	0.53
Sm <sub>2</sub> O <sub>3</sub>	1.13	1.58	1.50	0.73	0.73	0.54	0.78	0.75	1.55	0.74	1.68
Eu <sub>2</sub> O <sub>3</sub>	0.04	0.00	0.13	0.00	0.00	0.09	0.00	0.00	0.00	0.00	0.09
Gd <sub>2</sub> O <sub>3</sub>	3.66	4.34	4.28	2.48	2.46	2.46	2.83	2.36	4.08	2.67	4.70
Tb <sub>2</sub> O <sub>3</sub>	0.77	0.90	0.85	0.60	0.56	0.66	0.69	0.57	0.91	0.66	0.97
Dy <sub>2</sub> O <sub>3</sub>	6.10	6.42	6.31	4.97	5.11	5.20	5.48	4.88	6.02	5.12	6.85
Ho <sub>2</sub> O <sub>3</sub>	1.08	1.04	1.05	0.91	0.99	0.76	1.04	0.94	0.98	1.05	1.11
Er <sub>2</sub> O <sub>3</sub>	4.37	4.11	4.18	4.30	4.36	2.93	4.04	4.20	3.82	4.48	4.48
Tm <sub>2</sub> O <sub>3</sub>	0.72	0.62	0.64	0.79	0.78	0.45	0.65	0.66	0.63	0.77	0.69
Yb <sub>2</sub> O <sub>3</sub>	3.96	3.28	3.44	5.79	5.17	2.50	3.99	3.76	3.36	4.62	3.82
Lu <sub>2</sub> O <sub>3</sub>	0.60	0.47	0.56	1.07	0.84	0.41	0.48	0.49	0.35	0.54	0.63
CaO	0.07	0.11	0.03	0.02	0.10	0.17	0.01	0.03	0.16	0.05	0.01
FeO total	0.03	0.01	0.01	0.00	0.04	1.37	0.57	0.23	0.08	0.07	0.64
PbO	0.00	0.09	0.09	0.07	0.08	0.00	0.09	0.05	0.02	0.02	0.01
Total	99.88	101.26	99.73	99.81	99.85	101.22	100.65	100.21	100.63	100.24	101.20

1996; Broska et al., 2012<sup>b</sup>). This typomorphic distribution of accessory phases is most likely controlled by P, LREE and Ca content in melt and by magmatic differentiation and redox conditions.

The accessory mineral assemblage of major allanite-(Ce) + ilmenite ± magnetite in the Velence granite indicates intermediate oxygen fugacity ( $f_{O_2}$ ) values (cf. Uher & Broska, 1996), and the relatively high activity of Ca stabilises allanite-(Ce) while



**Fig. 9:** Chondrite-normalised REE patterns A – in xenotime-(Y) from parental granite and pegmatite/aplite, B – Detail of HREE tetrad effect. Normalised values after Barrat et al. (2012).

**Table 4:** continued - Representative compositions of EPMA of xenotime-(Y) (apfu).

End member	Xtm	Xtm	Xtm	Xtm	Xtm	Xtm	Xtm	Xtm	Xtm	Xtm	Xtm
S <sup>6+</sup>	0.000	0.001	0.000	0.001	0.000	0.003	0.000	0.001	0.000	0.000	0.000
P <sup>5+</sup>	0.973	0.956	0.953	0.992	0.988	0.959	0.946	0.955	0.917	0.983	0.947
As <sup>5+</sup>	0.000	0.000	0.000	0.000	0.001	0.002	0.001	0.002	0.001	0.001	0.000
Si <sup>4+</sup>	0.036	0.054	0.051	0.025	0.031	0.039	0.055	0.051	0.136	0.024	0.040
Th <sup>4+</sup>	0.006	0.020	0.019	0.008	0.006	0.001	0.017	0.008	0.001	0.003	0.004
U <sup>4+</sup>	0.006	0.012	0.014	0.009	0.013	0.001	0.011	0.008	0.005	0.004	0.008
Al <sup>3+</sup>	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.030	0.000	0.000
Y <sup>3+</sup>	0.726	0.690	0.696	0.714	0.712	0.803	0.744	0.770	0.675	0.758	0.720
La <sup>3+</sup>	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Ce <sup>3+</sup>	0.001	0.002	0.002	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001
Pr <sup>3+</sup>	0.003	0.003	0.002	0.003	0.002	0.002	0.002	0.002	0.002	0.002	0.002
Nd <sup>3+</sup>	0.005	0.008	0.009	0.004	0.006	0.001	0.003	0.004	0.006	0.002	0.006
Sm <sup>3+</sup>	0.013	0.019	0.018	0.009	0.008	0.006	0.009	0.009	0.017	0.009	0.020
Eu <sup>3+</sup>	0.001	0.000	0.002	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.001
Gd <sup>3+</sup>	0.041	0.049	0.049	0.028	0.028	0.026	0.032	0.026	0.044	0.030	0.053
Tb <sup>3+</sup>	0.009	0.010	0.010	0.007	0.006	0.007	0.008	0.006	0.010	0.007	0.011
Dy <sup>3+</sup>	0.067	0.070	0.071	0.054	0.056	0.054	0.060	0.053	0.063	0.055	0.076
Ho <sup>3+</sup>	0.012	0.011	0.012	0.010	0.011	0.008	0.011	0.010	0.010	0.011	0.012
Er <sup>3+</sup>	0.047	0.044	0.046	0.046	0.046	0.030	0.043	0.044	0.039	0.047	0.048
Tm <sup>3+</sup>	0.008	0.007	0.007	0.008	0.008	0.005	0.007	0.007	0.006	0.008	0.007
Yb <sup>3+</sup>	0.041	0.034	0.036	0.060	0.053	0.025	0.041	0.038	0.033	0.047	0.040
Lu <sup>3+</sup>	0.006	0.005	0.006	0.011	0.009	0.004	0.005	0.005	0.003	0.005	0.007
Ca <sup>2+</sup>	0.003	0.004	0.001	0.001	0.003	0.006	0.000	0.001	0.006	0.002	0.000
Fe <sup>2+</sup>	0.001	0.000	0.000	0.000	0.001	0.037	0.016	0.007	0.002	0.002	0.018
Pb <sup>2+</sup>	0.000	0.001	0.001	0.001	0.001	0.000	0.001	0.000	0.000	0.000	0.000
Position X	1.009	1.012	1.004	1.018	1.021	1.002	1.002	1.009	1.054	1.008	0.988
Position A	0.994	0.988	1.000	0.973	0.970	1.018	1.011	0.999	0.956	0.993	1.035
Total	2.003	2.000	2.004	1.990	1.991	2.020	2.013	2.008	2.010	2.001	2.023

monazite-(Ce) occurrence indicates relatively low Ca activity (Lee & Dodge, 1964; Lee & Silver, 1964; Lee & Bastron, 1967; Snetsinger, 1967; Cuney & Friedrich, 1987; Parrish, 1990; Casillas et al., 1995; Broska et al., 2006). Although allanite-(Ce) is present to the exclusion of monazite-(Ce) in rocks that contain more than about 2.0 wt.% CaO and only monazite-(Ce) is present in rocks with less than 0.7% CaO, both allanite-(Ce) and monazite-(Ce) are present at intermediate CaO values (Lee & Dodge, 1964; Lee & Bastron, 1967).

In contrast, monazite-(Ce) composition is normally almost the pure REE endmember when it is present in A-type granitoids, especially in hypersolvus conditions. This allows only limited ThSiREE<sub>1</sub>P<sub>1</sub> huttonite entry and/or negligible Ca(Th,U)REE<sub>2</sub> cheralite substitution. This appears to be a characteristic feature of hypersolvus A-type granitoid rocks where Ca is lacking in the bulk-rock (0.05 wt.% CaO) - (Ondrejka et al., 2007).

The MGM from the subsolvus Velence granitic rock samples are relatively rich in Th ( $\leq 10.6$  wt.% ThO<sub>2</sub>). This is most likely related to higher Th content in monazite-(Ce) which crystallized at higher temperatures (Broska et al., 2000), and it accords with the hot and dry origin of A-type granitic rocks

(cf. Whalen et al., 1987). In contrast, Th-enrichment is more developed in the pegmatite/aplite varieties and, combined with Ca, it stabilises cheralitic monazite and cheralite as the relatively common accessory phase (Gramaccioli & Segalstad, 1978; Bea, 1996; Förster, 1998; Pérez-Soba et al., 2014; Uher et al., 2014). Here, Ca-rich MGM most likely crystallised as an early magmatic or orthomagmatic phase before Ca was consumed by formation of primary allanite-(Ce) as the dominant REE accessory mineral.

While MGM is typically Ce-dominant in parental granite, there are some instances of La or Nd endmember dominance with Ce/La = 0.78 and Ce/Nd = 0.47. These monazite-(La) and monazite-(Nd) endmembers and enrichment trends towards La  $\approx$  Nd > Ce are only observed in MGM from the pegmatite samples. Here, negative Ce anomaly is recognised and such Ce deficiency relative to La and Nd cannot be therefore explained by selective and differential REE complexing in the granite-to-pegmatite solidification sequence where the tetrad effect on LREE is usually developed (e.g., Masau et al., 2002). Cerium anomalies in REE accessory minerals commonly develop in highly oxidised and fluid-rich environments, which causes fractionation of REE

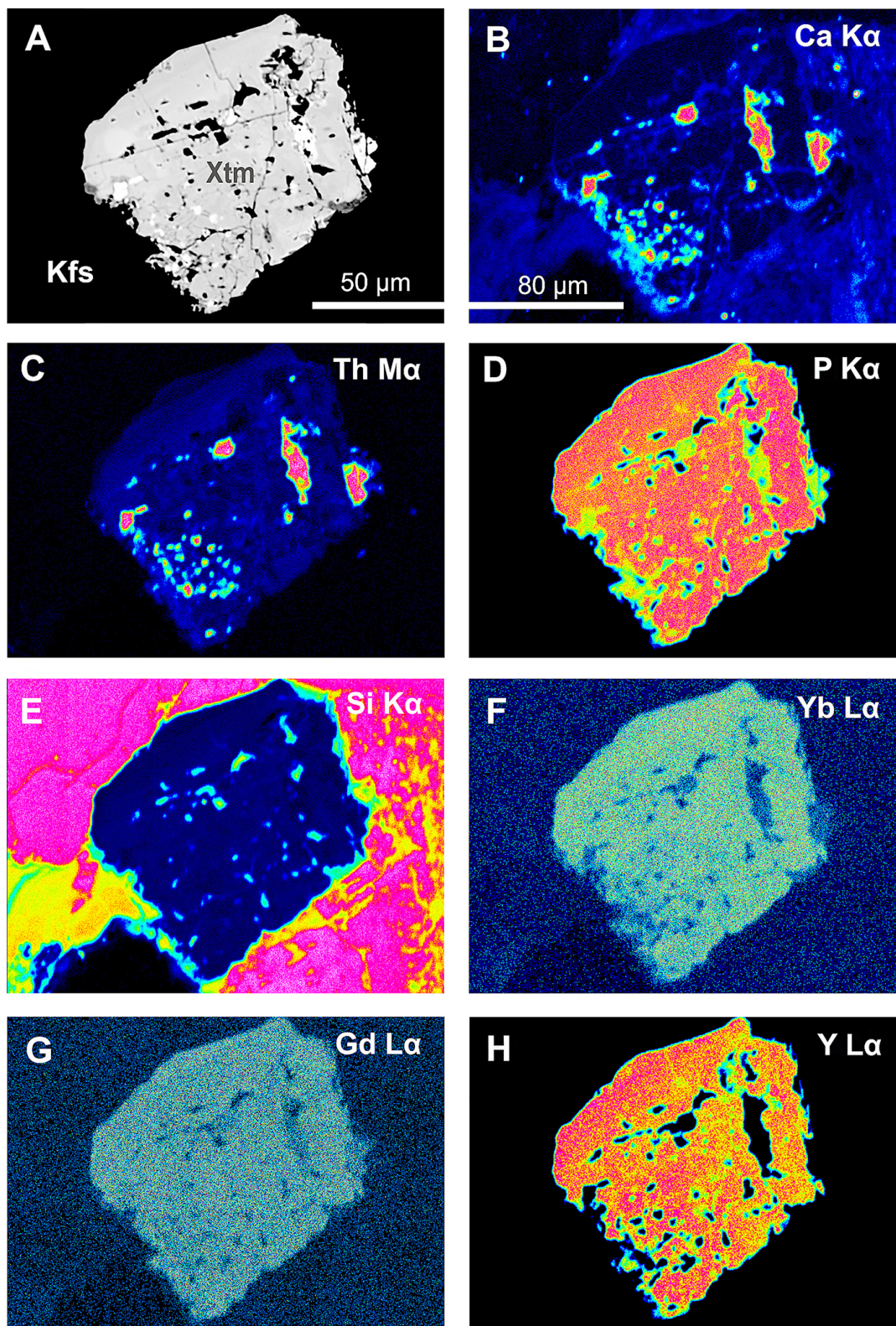
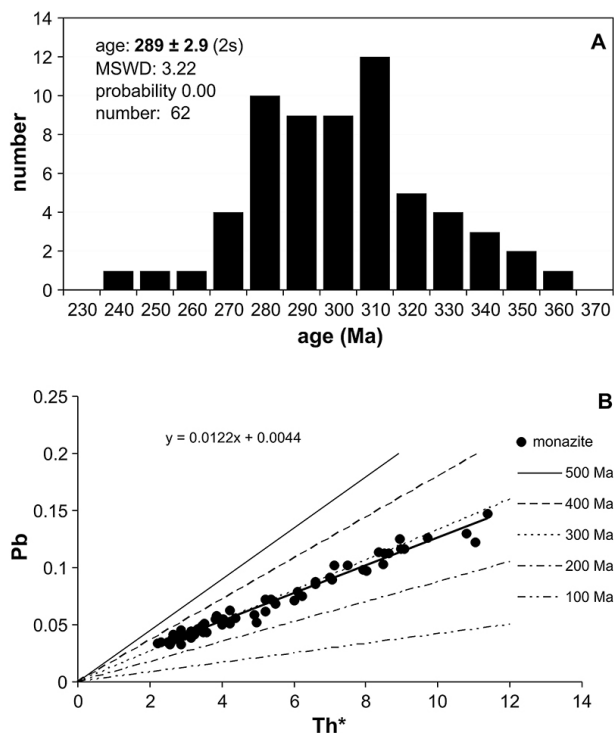


Fig. 10: False colour X-ray mapping showing the distribution of chemical elements in xenotime-(Y). False colours: blue – very low, green – low, yellow – medium, orange – high, red – very high.

by oxidation of  $Ce^{3+}$  to  $Ce^{4+}$  (Fisher & Meyrowitz, 1962; Banfield & Eggleton, 1989; Braun et al., 1990; Demartin et al., 1991<sup>b</sup>; Gieré & Sorensen, 2004; Mayer et al., 2014; MacDonald et al., 2015; Bagiński et al., 2016; Ondrejka et al., 2018). In contrast, weakly developed tetrad effect in T3 (Gd to Ho) and T4 (Er to Lu) tetrads in xenotime-(Y) is probably an inherited geochemical

signature of A-type parental melt (rich in F), since it occurs also in xenotime-(Y) from granite and gradually evolves during the progressive crystallization of the pegmatite derivatives. The mechanism of formation of the tetrad effect remains still unclear, but it is obviously connected to F-controlled complexing in highly evolved ligand-rich environments (e.g., Irber, 1999;

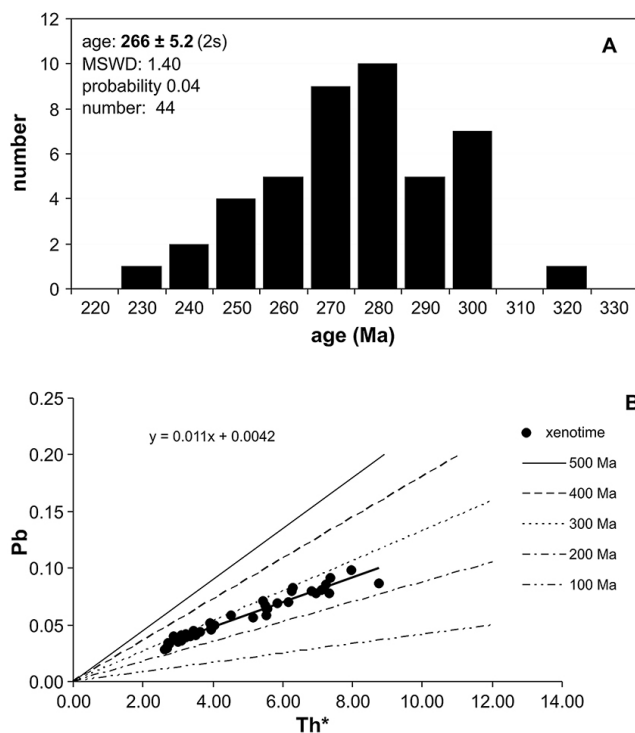


**Fig. 11:** Th–U–total Pb EPMA dating. A: histogram, B: isochron diagram of MGM ages from Velence. Pb values are in wt. %,  $Th^* = Th + 3.15 \cdot U$  wt. %.

Masau et al., 2002; Veksler et al., 2005; Badanina et al., 2006; Čopiaková et al., 2015).

Although, the concentration of fluorine in MGM from parental granite is negligible and often zero, an increased F content (0.3 – 1.2 wt.%) was observed in MGM from pegmatite. This suggests that F was preferentially consumed by simultaneous crystallization of primary fluorapatite (cf. Broska, 2001). Some large Ca–Th-rich crystals with low totals in pegmatite most likely represent altered(?) cheralitic MGM or brockitic rhabdophane group minerals (RGM) where elevated Y and HREE anomaly are typical compositional features (Nagy et al., 2002; Krenn & Finger, 2007; Ondrejka et al., 2018) and are in contrast with structural adjustments to the different sizes of REE atoms in monazite-(Ce) and xenotime-(Y) dimorphs (Ni et al., 1995). These brockite- and trisramite-rich supergene RGM with euhedral hexagonal symmetry associated with alunite supergroup minerals, clay minerals and goethite are also present in the Velence microgranite (Ondrejka et al., 2018).

While monazite-(Ce) from parental granite remains relatively unaffected by later post-magmatic-hydrothermal alteration, xenotime-(Y) was partly altered. This takes the form of remobilised  $CaTh(PO_4)_2$  and particularly  $(Th, U)SiO_4$  enrichment of a xenotime-(Y) portion via the coupled substitution reactions  $Ca^{2+} + Th^{4+} \leftrightarrow 2REE^{3+}$ , and  $(Th, U)^{4+} + Si^{4+} \leftrightarrow REE^{3+} + P^{5+}$  respectively (e.g., Harlov, 2010). The partial chemical alteration of xenotime-(Y) caused irregular patchy zoning and subsequent growth of numerous tiny cheralite and thorite inclusions close to altered areas (Fig. 2H, J), and this can be explained by the occurrence of a younger dissolution-reprecipitation process (e.g., Harlov et al., 2005; Putnis et al., 2007; Ondrejka et al.,



**Fig. 12:** Th–U–total Pb EPMA dating. A: histogram, B: isochron diagram of xenotime-(Y) ages from Velence. Pb values are in wt. %,  $Th^* = Th + 3.15 \cdot U$  wt. %.

2016). This required in-situ fluid-aided element redistribution; especially of Ca, Th, and U. A very similar dissolution-reprecipitation microtexture of thorite and/or uraninite inclusions in xenotime-(Y) and associated monazite-(Ce) has been described in granitic pegmatites in the Hidra anorthosite massif in Norway (Hetherington & Harlov, 2008) and in granitic orthogneisses in the Veporic pre-Alpine-basement, Western Carpathians, Slovakia (Ondrejka et al., 2016). Textural and crystal-chemical evidence therefore suggests that cheralite and thorite inclusions occurred from fluid-mineral interaction in a closed chemical system, where all components necessary for nucleation and growth of the inclusions were already present in the host xenotime-(Y) – (Hetherington & Harlov, 2008). This also suggest the relatively low mobility of actinides during dissolution-reprecipitation because significant amounts were unable to leave the xenotime-(Y) crystal (Ondrejka et al., 2012, 2016).

## 5.2. Monazite and xenotime formation and age

The presented  $289 \pm 2.9$  Ma EPMA monazite in-situ chemical age is within error of the  $283 \pm 5$  Ma; in-situ U–Pb SHRIMP zircon age (Uher & Ondrejka, 2009) from the same granitic rocks and indicates an Early Permian (Cisuralian) magmatic event. These ages are supported by previously published  $280 \pm 7$  Ma Rb–Sr whole-rock dating (Buda, 1985) and 271–291 Ma K–Ar, and Rb–Sr biotite dating (Horváth et al., 2004). These suggest older Velence granite massif age than similar A-type granite plutonism in the Western Carpathian region (Turčok, Hrončok, Upohlav: 262–267 Ma). This significant time gap of approximately 20 Ma

Table 5: Analytical data and ages of monazite-(Ce) from the investigated samples.

Sample	Th	U	Pb	Y	Age	Age	Sample	Th	U	Pb	Y	Age	Age
	wt.%	wt.%	wt.%	wt.%	(Ma)	2 $\sigma$		wt.%	wt.%	wt.%	wt.%	(Ma)	2 $\sigma$
VEL-1	5.340	0.050	0.069	0.879	280	23.1	VEL-2	2.421	0.167	0.041	1.897	309	42.7
VEL-1	3.660	0.059	0.057	1.039	333	32.1	VEL-2	2.633	0.160	0.039	1.906	275	40.2
VEL-1	3.114	0.041	0.041	0.963	283	37.6	VEL-2	1.913	0.118	0.034	1.713	334	54.9
VEL-1	2.370	0.037	0.034	1.154	309	49.0	VEL-2	3.419	0.245	0.063	2.193	333	30.7
VEL-1	3.873	0.054	0.054	1.598	301	31.0	VEL-2	4.891	0.096	0.072	1.231	310	25.3
VEL-1	3.876	0.038	0.055	0.534	306	31.0	VEL-2	4.922	0.088	0.062	1.218	267	25.0
VEL-1	3.071	0.026	0.044	0.963	312	38.7	VEL-2	4.998	0.112	0.072	1.268	302	24.5
VEL-1	3.338	0.035	0.049	0.898	319	35.4	VEL-2	7.881	0.236	0.113	1.683	292	16.1
VEL-1	2.690	0.035	0.038	0.769	300	43.6	VEL-2	9.638	0.362	0.130	2.987	270	13.5
VEL-1	3.398	0.024	0.047	0.645	305	35.2	VEL-2	6.935	0.332	0.097	3.225	273	17.4
VEL-1	2.474	0.038	0.034	0.860	295	45.9	VEL-3	8.000	0.160	0.113	0.922	296	16.0
VEL-1	2.742	0.041	0.033	0.994	255	43.0	VEL-3	8.034	0.134	0.103	1.159	273	16.1
VEL-1	4.053	0.052	0.051	0.640	269	29.3	VEL-3	6.993	0.151	0.102	1.269	305	18.0
VEL-1	3.459	0.035	0.043	1.024	271	34.7	VEL-3	4.182	0.061	0.056	0.794	286	28.6
VEL-1	4.753	0.042	0.059	0.651	270	25.4	VEL-3	4.655	0.094	0.052	1.604	236	25.3
VEL-1	3.108	0.060	0.045	0.867	307	37.5	VEL-3	3.135	0.043	0.044	0.691	300	37.8
VEL-1	2.412	0.050	0.033	1.315	287	49.2	VEL-3	2.577	0.051	0.038	0.689	307	44.6
VEL-1	6.956	0.018	0.091	1.070	291	19.2	VEL-3	3.365	0.037	0.043	0.617	277	35.1
VEL-1	2.182	0.016	0.034	1.001	341	56.0	VEL-3	4.903	0.170	0.070	1.050	290	22.5
VEL-1	2.556	0.021	0.037	1.035	313	48.2	VEL-3	5.493	0.192	0.079	1.026	289	20.0
VEL-1	6.520	0.029	0.086	1.091	290	20.2	VEL-3	8.423	0.202	0.116	0.731	287	14.1
VEL-1	2.557	0.017	0.038	0.831	324	48.5	VEL-3	10.575	0.244	0.147	0.706	291	11.5
VEL-1	2.568	0.025	0.041	0.881	347	47.2	VEL-3	8.935	0.245	0.126	0.358	290	13.3
VEL-1	3.428	0.027	0.051	0.541	324	36.0	VEL-3	3.617	0.061	0.055	0.647	322	31.7
VEL-2	4.866	0.424	0.075	3.023	270	21.5	VEL-3	2.766	0.033	0.041	0.621	316	41.5
VEL-2	5.846	0.229	0.087	2.665	296	20.6	VEL-3	6.363	0.220	0.090	1.284	284	18.0
VEL-2	5.247	0.239	0.071	3.099	264	22.3	VEL-8	7.970	0.119	0.113	0.437	304	15.6
VEL-2	10.197	0.265	0.122	2.181	248	12.8	VEL-8	7.554	0.116	0.098	0.411	277	16.3
VEL-2	1.680	0.511	0.046	1.819	308	38.1	VEL-8	8.310	0.197	0.125	0.552	312	14.6
VEL-2	2.701	0.055	0.045	0.532	351	39.8	VEL-8	4.943	0.672	0.102	2.452	321	17.8
VEL-2	3.452	0.175	0.050	1.830	279	31.9	VEL-8	7.169	0.554	0.116	1.896	291	14.7

between Velence granite magmatic crystallization and other Western Carpathians A-type occurrences most likely reflects different palaeogeographic position, and therefore different timing of this Permian granite.

The presented xenotime-(Y) in-situ chemical dating reveals post-Variscan age corresponding to the Middle Permian (Gadalupean) stage. However, it is systematically younger ( $\approx 20$  Ma) than magmatic monazite-(Ce) in the host granite. The most plausible interpretations of this age discrepancy are: (1) the xenotime-(Y) age indicates younger, late-magmatic crystallization than the early-magmatic age of monazite-(Ce) precipitation; or (2) post-magmatic (subsolvus) recrystallization of orthomagmatic xenotime-(Y) due to younger event and subsequent overprint of the parental granitic rocks. The age difference between monazite-(Ce) and xenotime-(Y) EPMA chemical ages for the same granite

( $\approx 20$  Ma) is generally too great, thus the second scenario is most likely. Moreover, most xenotime-(Y) crystals show partial alteration and formation of secondary microtexture characterised with occurrence of Th-silicate (thorite?) and/or cheralite inclusions (Fig. 2H, J) and irregular zoning (Fig. 2L). This most likely reflects the dissolution-reprecipitation process (check chapter 5.1.) herein, and connection with rejuvenation. In addition, the  $266 \pm 5.2$  Ma xenotime-(Y) EPMA age is clearly comparable to SHRIMP zircon ages ( $262 \pm 4$  Ma –  $267 \pm 2$  Ma) for the Western Carpathian A-type granite occurrences (Turčok, Hrončok and Upohlav; Ondrejka et al., unpubl.). This further suggests that fluid-assisted xenotime-(Y) recrystallization is connected with increased heat transfer during the granites emplacement.

Finally, the rare presence of ca. 670 – 640 Ma old monazite-(Ce) domains indicates admixture of Neoproterozoic material

**Table 6: Analytical data and ages of xenotime-(Y) from the investigated samples.**

Sample	Th	U	Pb	Y	Age	Age	Sample	Th	U	Pb	Y	Age	Age
	wt.%	wt.%	wt.%	wt.%	(Ma)	2 $\sigma$		wt.%	wt.%	wt.%	wt.%	(Ma)	2 $\sigma$
VEL-1	0.617	0.629	0.029	32.881	248	62.2	VEL-2F	0.970	1.093	0.058	31.239	292	38.1
VEL-1	1.275	1.316	0.064	30.444	261	30.4	VEL-2F	1.084	1.376	0.059	31.078	240	31.5
VEL-1	1.258	1.517	0.070	31.263	257	27.3	VEL-2F	0.735	1.577	0.070	31.226	269	29.5
VEL-1	0.693	0.745	0.037	31.632	271	53.5	VEL-2F	1.666	1.758	0.078	30.922	240	23.9
VEL-1	0.728	0.810	0.040	31.856	268	48.7	VEL-2F	1.688	1.749	0.092	31.148	281	24.1
VEL-1	0.601	0.648	0.029	31.479	244	60.6	VEL-8A	0.785	0.850	0.043	41.800	274	46.1
VEL-1	0.618	0.769	0.041	32.300	298	53.3	VEL-8A	1.896	1.353	0.083	41.207	297	26.7
VEL-1	0.556	0.666	0.034	31.067	282	60.6	VEL-8A	1.116	0.876	0.045	41.436	260	41.4
VEL-1	2.097	1.552	0.081	29.645	256	24.1	VEL-8C	0.039	0.575	0.705	41.737	317	56.8
VEL-1	1.577	1.213	0.067	30.553	273	30.8	VEL-8C	0.043	0.678	0.852	45.819	292	47.4
VEL-1	0.438	0.289	0.728	32.484	299	117.2	VEL-8B	0.035	0.654	0.757	43.132	263	52.6
VEL-1	0.724	0.705	0.038	31.621	288	54.2	VEL-8B	0.040	0.741	0.854	42.006	264	46.6
VEL-1	0.980	1.292	0.056	30.635	247	32.3	VEL-8B	0.041	0.529	0.833	41.404	295	50.4
VEL-1	1.644	1.731	0.086	29.615	267	23.4	VEL-8B	0.038	0.520	0.817	41.759	284	51.5
VEL-1	1.146	1.364	0.064	31.581	260	30.0	VEL-8B	0.041	0.258	0.992	45.096	272	47.0
VEL-1	1.155	1.324	0.071	30.240	293	30.8	VEL-8B	0.035	0.500	0.762	42.896	277	54.9
VEL-1	2.491	1.692	0.098	29.272	278	21.3	VEL-8B	0.050	0.459	1.065	45.615	297	41.7
VEL-1	0.768	0.881	0.044	31.938	275	46.3	VEL-8B	0.080	1.079	1.601	44.612	286	26.8
VEL-1	0.485	0.662	0.029	32.034	247	64.5	VEL-8B	0.037	0.865	0.731	43.967	269	50.1
VEL-1	2.246	1.459	0.077	30.045	251	25.2	VEL-8B	0.033	0.820	0.677	43.447	264	54.2
VEL-1	0.617	0.658	0.033	31.360	271	61.0	VEL-8B	0.048	0.960	0.954	43.210	275	40.3
VEL-2F	1.325	1.697	0.080	31.053	264	25.8	VEL-8B	0.047	0.970	0.924	43.004	272	41.2

in the granite protolith. This systematic presence of an older recycled material is also supported by the occurrence of 650 – 630 Ma old inherited zircon cores in the Velence granite dated by SHRIMP method (cf. Uher & Ondrejka, 2009).

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