



# Description and recognition of potassic-richterite, an amphibole supergroup mineral from the Pajsberg ore field, Värmland, Sweden

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## Abstract

Potassic-richterite, ideally  ${}^A\text{K}^B(\text{NaCa})^C\text{Mg}_5\text{Si}_8\text{O}_{22}^W(\text{OH})_2$ , is recognized as a valid member of the amphibole supergroup (IMA-CNMNC 2017–102). Type material is from the Pajsberg Mn-Fe ore field, Filipstad, Värmland, Sweden, where the mineral occurs in a Mn-rich skarn, closely associated with mainly phlogopite, jacobsonite and tephroite. The megascopic colour is straw yellow to grayish brown and the luster vitreous. The nearly anhedral crystals, up to 4 mm in length, are pale yellow (non-pleochroic) in thin section and optically biaxial (–), with  $\alpha = 1.615(5)$ ,  $\beta = 1.625(5)$ ,  $\gamma = 1.635(5)$ . The calculated density is  $3.07 \text{ g}\cdot\text{cm}^{-3}$ .  $\text{VHN}_{100}$  is in the range 610–946. Cleavage is perfect along  $\{110\}$ . EPMA analysis in combination with Mössbauer and infrared spectroscopy yields the empirical formula  $(\text{K}_{0.61}\text{Na}_{0.30}\text{Pb}_{0.02})_{\Sigma 0.93}(\text{Na}_{1.14}\text{Ca}_{0.79}\text{Mn}_{0.07})_{\Sigma 2}(\text{Mg}_{4.31}\text{Mn}_{0.47}\text{Fe}^{3+}_{0.20})_{\Sigma 5}(\text{Si}_{7.95}\text{Al}_{0.04}\text{Fe}^{3+}_{0.01})_{\Sigma 8}\text{O}_{22}(\text{OH}_{1.82}\text{F}_{0.18})_{\Sigma 2}$  for a fragment used for collection of single-crystal X-ray diffraction data. The infra-red spectra show absorption bands at  $3672 \text{ cm}^{-1}$  and  $3736 \text{ cm}^{-1}$  for the  $\alpha$  direction. The crystal structure was refined in space group  $C2/m$  to  $R1 = 3.6\%$  [ $I > 2\sigma(I)$ ], with resulting cell parameters  $a = 9.9977(3) \text{ \AA}$ ,  $b = 18.0409(4) \text{ \AA}$ ,  $c = 5.2794(2) \text{ \AA}$ ,  $\gamma = 104.465(4)^\circ$ ,  $V = 922.05(5) \text{ \AA}^3$  and  $Z = 2$ . The  $A$  and  $M(4)$  sites split into  $A(m)$  ( $\text{K}^+$ ),  $A(2/m)$  ( $\text{Na}^+$ ),  $A(2)$  ( $\text{Pb}^{2+}$ ), and  $M(4')$  ( $\text{Mn}^{2+}$ ) subsites, respectively. The remaining  $\text{Mn}^{2+}$  is strongly ordered at the octahedrally coordinated  $M(2)$  site, possibly together with most of  $\text{Fe}^{3+}$ . The skarn bearing potassic-richterite formed at peak metamorphism, under conditions of low  $\text{SiO}_2$  and  $\text{Al}_2\text{O}_3$  activities and relatively high oxygen fugacities.

**Keywords** Potassic-richterite · Amphibole supergroup · Crystal structure · Skarn · Pajsberg · Långban-type deposit · Sweden

## Introduction

Potassic-richterite [ideally  $\text{K}(\text{NaCa})\text{Mg}_5\text{Si}_8\text{O}_{22}(\text{OH})_2$ , derived from the root species richterite via the homovalent substitution  ${}^{[A]}\text{Na}^+ \rightarrow {}^{[A]}\text{K}^+$ ], is not a new mineral in the sense that amphibole of this composition never has been reported before; on the contrary, already in the 19<sup>th</sup> and early 20<sup>th</sup> centuries, published wet-chemical analyses indicated the existence of K-

dominant amphibole varieties in what is now defined as the richterite compositional space. The analysed specimens from this period were mainly from the Långban Mn-Fe deposit, Sweden (Magnusson 1930; Sundius 1945), the type locality for richterite. There are also plenty of results from the microprobe era proving amphibole compositions approaching potassic end-member composition. Identified samples of potassic-richterite (commonly referred to as “potassium richterite” or “K-richterite” in the literature) are typically from ultrapotassic mantle-derived rocks such as peridotite xenoliths (in kimberlite) and lamproites (e.g. Erlank and Finger 1970; Dawson and Smith 1982; Wagner and Velde 1986; Mitchell 2012). The species was also reported from the metamorphosed Praborna manganese ore deposit, Val d’Aosta, Italy (Mottana and Griffin 1986; specimen later studied by single-crystal X-ray diffraction by Oberti et al. 1992). A Sr-bearing potassic-richterite from the Murun alkaline Massif (Eastern-Siberian Region, Russia) was structurally characterized by Sokolova et al. (2000).

The F-analogue, potassic-fluoro-richterite, was described by Della Ventura et al. (1992) from a volcanic ejectum from

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a pyroclastic deposit, Monte Somma, Italy. A K-rich richteritic amphibole with  $\text{Fe}^{2+} > \text{Mg}$  (“potassic-ferro-richterite”) is indicated by some analyses (e.g. Hogarth 1997), but is not yet approved. Potassic-richterite is not to be confused with “KK-richterite”, a synthetic ultrahigh-pressure amphibole of composition  $\text{K}(\text{KCa})\text{Mg}_5\text{Si}_8\text{O}_{22}(\text{OH})_2$  (e.g. Yang et al. 1999). Mazdab (2003) reviewed the diversity of K-dominant amphiboles and pointed out the need for complete characterization to achieve species status for some members.

The present paper presents data for the designated holotype specimen of potassic-richterite, approved as a valid species by the International Mineralogical Association-Commission on New Minerals, Nomenclature and Classification (IMA-CNMNC 2017–102); the name complies with the current IMA nomenclature for the amphibole supergroup (Hawthorne et al. 2012). Holotype material is deposited in the mineral collections of the Department of Geosciences, Swedish Museum of Natural History (GEO-NRM), Box 50007, SE-10405 Stockholm, Sweden, under collection number 19311387.

## Description and characterisation of potassic-richterite

### Occurrence and paragenesis

On the original label of the type specimen, it is written “Pajsberg iron and manganese ore mines”, which could either mean the Stora Pajsberg mine ( $59^\circ 47.04' \text{ N}$ ;  $14^\circ 19.03' \text{ E}$ ), or the Harstigen mine ( $59^\circ 47.10' \text{ N}$ ,  $14^\circ 18.84' \text{ E}$ ) 200 m to the NW. Both mines belong to the Pajsberg ore field near the northernmost tip of Lake Yngen (200 m a.s.l.), Filipstad, Värmland, Sweden. However, the present sample shows the greatest physical resemblance to skarn from the Stora Pajsberg mine. It was originally collected by the mineralogist Lars Johan Igelström, probably in the 1850s, and later acquired by the chemist Johan Fredrik Lundberg. It was finally incorporated in the mineral collection of the Swedish Museum of Natural History in 1931, as “manganophyllite (manganous phlogopite) with richterite”. The occurrence of “richterite” from Pajsberg was first reported by Igelström (1867), as a common component of Mn-skarn associated with tephroite and phlogopite. Igelström’s original chemical analyses of amphibole were of a semi-quantitative character, with only the sum of alkalis ( $\text{Na}_2\text{O} + \text{K}_2\text{O}$ ) given.

The mines are located in Långban-type Mn-Fe oxide deposits hosted by dolomitic marble, with spatially separated Mn (hausmannite) and Fe (hematite-magnetite) ores, surrounded by dominantly felsic metavolcanic and metavolcaniclastic rocks. According to Hamberg (1890), richterite belongs to a group of earliest formed skarn minerals (olivine, garnet, pyroxene, rhodonite, amphibole) at the

Harstigen deposit, followed by various Mn, Pb, Ba, As, Sb, and Be minerals (hydrous silicates, carbonates, sulphates, arsenates, oxyhalides, sulphides, native elements) formed in fissures and cavities at lower temperatures. Generally, the Paleoproterozoic Långban-type deposits in the Bergslagen ore region originally deposited as chemical precipitates from volcanogenic hydrothermal solutions in a shallow submarine environment (Boström et al. 1979; Holtstam and Mansfeld 2001). Subsequent episodes of hydrothermal alteration, metamorphism and deformation spanning 1.9–1.0 Ga led to the present-day skarn and late-stage mineral assemblages.

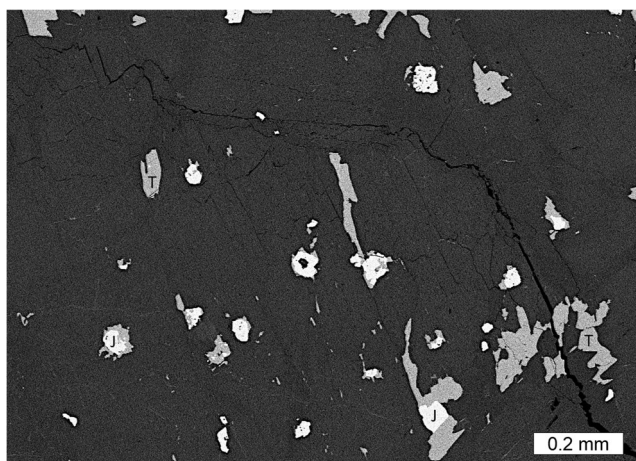
In the sample under study (Fig. 1), potassic-richterite occurs with manganous tetraferrian phlogopite, magnesian tephroite, jacobsite and minor oxyplumboroméite in a compact, Mn-rich skarn, as masses of randomly arranged crystals. About  $\frac{1}{4}$  by volume of the specimen, ca  $8 \times 7 \times 5$  cm in size, constitutes the amphibole, which shows a relatively sharp transition to a more fine-grained matrix of mainly phlogopite, jacobsite and minor calcite. Tephroite (ca 85 mol%  $\text{Mn}_2\text{SiO}_4$  according to energy dispersive X-ray microanalysis) forms irregular, sometimes elongated grains up to 0.5 mm that have partly developed along [001] of the amphibole, and jacobsite ( $\text{Mn}^{2+}[\text{Fe}^{3+}_{1.76}\text{Mn}^{3+}_{0.24}]\text{O}_4$  from microprobe data) occurs as mostly equigranular inclusions of up to 100  $\mu\text{m}$  in diameter (Fig. 2).

### Appearance and physical properties

Potassic-richterite exhibits a short-prismatic morphology; the crystals are subhedral to anhedral and commonly elongated along [001] with average crystal size of 1–2 mm, exceptionally up to 4 mm long. The crystals are vitreous translucent, with a straw yellow to brown colour, and a greyish white streak. In thin (ca 50  $\mu\text{m}$ ) crystal sections, potassic-richterite is transparent and pale yellow with no visible pleochroism. No fluorescence effects have been observed under ultraviolet light. Optically the mineral is biaxial negative, with



**Fig. 1** Image of potassic-richterite with jacobsite (black, granular). Sample GEO-NRM #1931137



**Fig. 2** Field-emission BSE image of potassic-richterite and associated minerals (J = jacobsonite, T = tephroite)

$\alpha = 1.615(5)$   $\beta = 1.625(5)$   $\gamma = 1.635(5)$  (white light) and  $2V_{\text{meas}} = 75(5)^\circ$  and  $Z \wedge c \sim 24^\circ$ . Dispersion is weak,  $r < v$ .

Potassic-richterite is brittle, with measured Vickers hardness number ( $VHN_{100}$ ) between 610 and 946 (average 806;  $n = 10$ ) corresponding to a Mohs hardness of 5–6, and has an uneven, splintery fracture. Cleavage is perfect along  $\{110\}$ . The density could not be measured directly due to the foreign mineral inclusions in the crystals. The calculated value is  $3.075 \text{ g}\cdot\text{cm}^{-3}$  (ideal formula and unit-cell parameters from X-ray single-crystal data), or  $3.070 \text{ g}\cdot\text{cm}^{-3}$  (empirical formula and unit-cell parameters from powder X-ray diffraction data). The Gladstone-Dale compatibility (Mandarino 1981) is  $1-(K_P/K_C) = 0.016$  (superior category).

## Chemical composition

The chemical composition of potassic-richterite was first determined from point analyses on a polished surface using an ARL-SEMQ electron probe micro-analyser (EPMA) in wavelength dispersive (WDS) mode (15 kV, 12 nA, beam size 1–2  $\mu\text{m}$ ), with calibrant materials: (Na, Ca Mg, Al, Si) hornblende-type melt glass, K orthoclase, Fe hematite, Mn rhodonite, F topaz. A second analysis was done with a JEOL 8200 Super Probe (WDS mode, 15 kV, 5 nA, 1  $\mu\text{m}$  beam) with calibrant materials: F hornblende, Sr celestine, Mn rhodonite, K orthoclase, Pb galena, Fe fayalite, (Ca, Si, Al) grossular, Cr metal, Ni nickeline, Ti ilmenite, Mg olivine, Ba sanbornite on the fragment used for the single-crystal study. Results are given in Table 1.

The presence of  $(\text{OH})^-$  is confirmed by Fourier-transform infrared (FTIR) spectroscopy and crystal-structure refinement, see below. Complementary analysis of other light elements (Li, B, F) was done by nuclear-reaction analysis at the proton-beamline facility (LIBAF) at Lund University, Sweden (Nilsson et al. 2017; De La Rosa et al. 2017). The results (in ppm) are: Li  $18 \pm 4$ , B  $104 \pm 6$  and F  $6440 \pm 480$ .

The average empirical formula based on 24 (O + OH + F) for the crystal fragment used in structure refinement is  $(\text{K}_{0.61}\text{Na}_{0.30}\text{Pb}_{0.02})_{\Sigma 0.93}(\text{Na}_{1.14}\text{Ca}_{0.79}\text{Mn}_{0.07})_{\Sigma 2}(\text{Mg}_{4.31}\text{Mn}_{0.47}\text{Fe}^{3+}_{0.20})_{\Sigma 5}(\text{Si}_{7.95}\text{Al}_{0.04}\text{Fe}^{3+}_{0.01})_{\Sigma 8}\text{O}_{22}(\text{OH}_{1.82}\text{F}_{0.18})_{\Sigma 2}$ , where essential components are in bold letters. The ideal formula is  ${}^A\text{K}^B(\text{NaCa})^C\text{Mg}_5^T\text{Si}_8\text{O}_{22}^W(\text{OH})_2$ , which requires  $\text{K}_2\text{O}$  5.65%,  $\text{Na}_2\text{O}$  3.71%,  $\text{CaO}$  6.72%,  $\text{MgO}$  24.15%,  $\text{SiO}_2$  57.60,  $\text{H}_2\text{O}$  2.16, total 100.0 wt.%.

On back-scattered electron (BSE) images, with maximum contrast, a slight heterogeneity in the composition of the amphibole is indicated (Fig. 2). This is in part related to variations in the Mn and Na concentrations, also notable if the two sets of analyses in Table 1 are compared. To certify that the specimen is K-dominant throughout, EDS point analyses (20 kV,  $n = 16$ ), were collected on different parts of six grains of potassic-richterite over an  $8 \times 10 \text{ mm}$  area in the polished section of the type specimen. A wider concentration range of  $\text{K}_2\text{O}$  is observed, 3.0–3.8 wt%, but with an average value of 3.6 wt%, comparable to the WDS data (Table 1). PbO was not included in the original microprobe analysis, but occurs in variable amounts, corresponding to up to 0.02 atoms per formula unit (pfu). F is the most variable element, possibly related to analytical difficulties, but it is clearly established that  $\text{OH} > \text{F}$  in this sample. Li and B are only present in trace amounts.

## Infrared spectroscopy and Mössbauer spectroscopy

A FTIR spectrum was collected on potassic-richterite powder (3 mg, mixed with KBr) in the range  $600\text{--}4000 \text{ cm}^{-1}$  at a spectral resolution of  $2 \text{ cm}^{-1}$  with a Bruker Vertex 70 spectrometer attached to a Bruker Hyperion 2000 IR-microscope. In the  $600\text{--}1500 \text{ cm}^{-1}$  range (Fig. 3) absorption bands are identified at  $1145 \text{ cm}^{-1}$ ,  $1077 \text{ cm}^{-1}$ ,  $1043 \text{ cm}^{-1}$ ,  $970 \text{ cm}^{-1}$ ,  $957 \text{ cm}^{-1}$ ,  $922 \text{ cm}^{-1}$ ,  $740 \text{ cm}^{-1}$ ,  $703 \text{ cm}^{-1}$ , and  $677 \text{ cm}^{-1}$ . These values are very similar to infrared (IR) data presented for potassic-richterite from the Murun Massif (Chukanov 2014).

Polarised single-crystal FTIR spectra in the range  $2000\text{--}8000 \text{ cm}^{-1}$  were collected from a  $56 \mu\text{m}$  thick doubly polished single-crystal fragment oriented parallel to (010), with the same instrument as mentioned above. The spectra (Fig. 4) show distinct absorption bands (with some shoulder features) at  $3672 \text{ cm}^{-1}$  and  $3736 \text{ cm}^{-1}$  in the  $\alpha$  direction caused by O-H stretching vibrations of the OH dipole at the O(3) site. The two main bands represent the  ${}^A\text{MgMgMg-SiSi}$  and  ${}^A\text{K-MgMgMg-SiSi}$  arrangements, respectively, observed for nominal end-member synthetic potassic-richterite (Hawthorne et al. 1997; Hawthorne and Della Ventura 2007). Mottana and Griffin (1986) reported bands at similar wavenumbers for two samples of natural potassic-richterite,  $3670 \text{ cm}^{-1}$  and  $3730 \text{ cm}^{-1}$ . The band at  $3736 \text{ cm}^{-1}$  shows a minor shoulder at  $3730 \text{ cm}^{-1}$  that is surely related to the  ${}^A\text{Na-MgMgMg-SiSi}$  arrangement. Less evident is the extending asymmetry toward lower wavenumbers

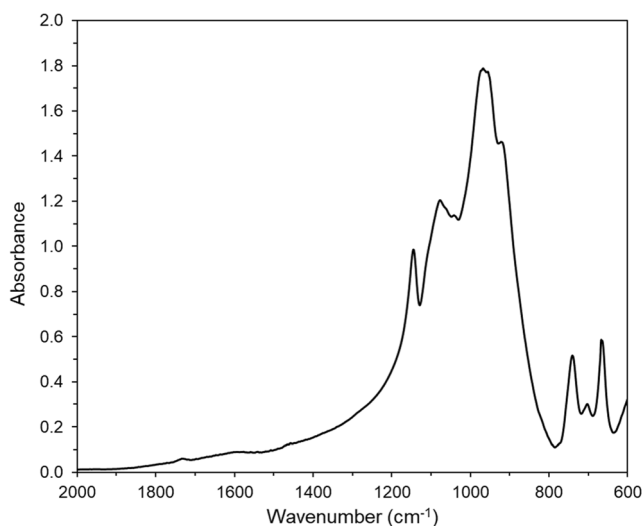
**Table 1** Results from EMPA analyses of potassic-richterite, with calculated cation proportions

Point*	0 <i>n</i> = 3	1	2	3	4	5	6	Average 1–6
EMPA results (wt%):								
SiO <sub>2</sub>	55.99	56.42	55.78	56.27	56.44	55.98	55.31	56.03
Al <sub>2</sub> O <sub>3</sub>	0.39	0.20	0.24	0.21	0.21	0.23	0.29	0.23
TiO <sub>2</sub>	0.00	0.00	0.04	0.00	0.02	0.02	0.00	0.01
Cr <sub>2</sub> O <sub>3</sub>	0.00	0.00	0.03	0.03	0.03	0.03	0.03	0.03
Fe <sub>2</sub> O <sub>3</sub>	1.65	2.10	2.03	1.98	1.72	1.97	2.11	1.99
MnO	5.22	4.88	4.79	4.21	3.95	4.51	4.51	4.48
NiO		0.00	0.01	0.01	0.00	0.03	0.08	0.02
MgO	20.92	20.09	20.08	20.51	20.61	20.50	20.39	20.36
CaO	5.52	5.04	5.06	5.17	5.52	5.15	5.10	5.17
SrO		0.04	0.11	0.01	0.05	0.00	0.00	0.04
BaO		0.05	0.03	0.00	0.00	0.05	0.00	0.02
Na <sub>2</sub> O	4.98	5.41	5.45	5.44	4.93	5.26	4.93	5.24
K <sub>2</sub> O	3.42	3.49	3.44	3.27	3.17	3.41	3.37	3.36
PbO		0.49	0.48	0.63	0.67	0.77	0.52	0.59
F	0.80	0.36	0.32	0.37	0.36	0.48	0.51	0.40
H <sub>2</sub> O*	1.74	1.95	1.95	1.94	1.95	1.89	1.85	1.92
Total	100.63	100.53	99.89	100.06	99.64	100.28	99.05	99.91
O=F	0.34	0.15	0.14	0.16	0.15	0.20	0.21	0.17
Total including O=F	100.29	100.37	99.75	99.91	99.49	100.07	98.84	99.74
Calculated mineral formulae (apfu):*								
Si	7.902	7.98	7.94	7.97	8.00	7.94	7.91	7.96
Al	0.06	0.02	0.04	0.03	0.00	0.04	0.05	0.04
Total T-site cations	7.97	8.00	7.98	8.00	8.00	7.98	7.96	7.99
Ti <sup>4+</sup>	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Al <sup>VI</sup>	0.00	0.01	0.00	0.01	0.04	0.00	0.00	0.00
Cr <sup>3+</sup>	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Fe <sup>3+</sup>	0.18	0.22	0.22	0.21	0.18	0.21	0.23	0.21
Mn <sup>2+</sup>	0.62	0.58	0.58	0.50	0.47	0.54	0.55	0.54
Mg	4.40	4.23	4.26	4.33	4.35	4.33	4.35	4.31
Ni <sup>2+</sup>	0.00	0.00	0.01	0.01	0.00	0.02	0.05	0.01
Total C-site cations	5.20	5.05	5.08	5.06	5.06	5.11	5.19	5.08
Ca	0.83	0.76	0.77	0.78	0.84	0.78	0.78	0.79
Na	1.36	1.48	1.50	1.49	1.35	1.45	1.37	1.44
K	0.62	0.63	0.62	0.59	0.57	0.62	0.62	0.61
Pb		0.02	0.02	0.02	0.03	0.03	0.02	0.02
Ba	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Sr	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00
Total A, B-site cations	3.01	2.90	2.93	2.89	2.80	2.88	2.78	2.86
F	0.36	0.16	0.15	0.17	0.16	0.21	0.23	0.18
OH	1.64	1.84	1.85	1.83	1.85	1.79	1.77	1.82
Total W-site anions	2.00	2.00	2.00	2.00	2.00	2.00	2.00	2.00

\*Point #0 is the average of three older EMP analyses. Points #1–6 represent analyses of the fragment used for single-crystal XRD study

\*\*Calculated based on 24(O,OH,F) apfu (atoms per formula unit)

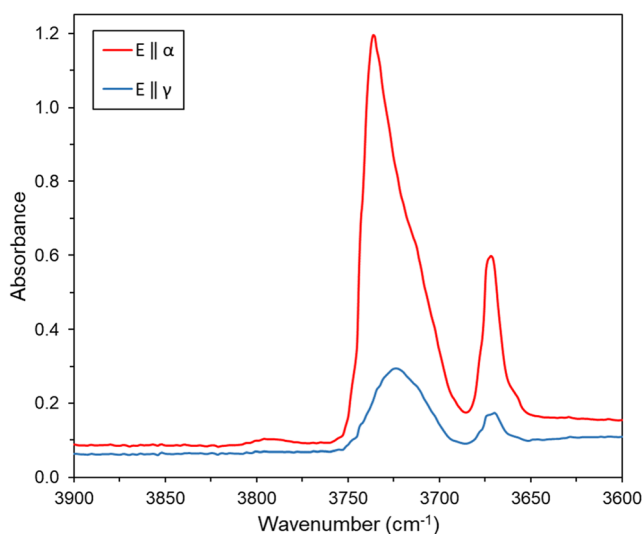




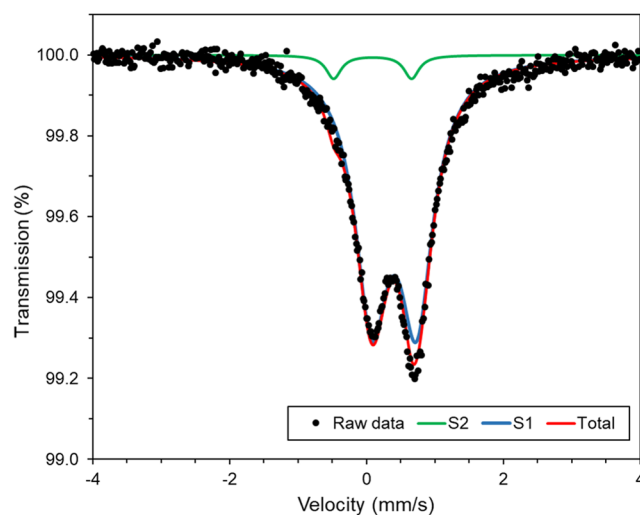
**Fig. 3** Powder FTIR spectrum of potassic-richterite for the 600–2000  $\text{cm}^{-1}$  range

that should be related to the  $^{\text{A}}\text{K-MgMgMg-SiAl}$  and  $^{\text{A}}\text{Na-MgMgMg-SiAl}$  configurations (Hawthorne and Della Ventura 2007), in accordance with the crystal chemistry ( $< 2\% \text{ } ^{\text{T}}\text{Al}$ ), see discussion below.

A sample of potassic-richterite was analyzed by Mössbauer spectroscopy using a conventional spectrometer system operated in constant-acceleration mode. Due to limited amounts of pure material, a  $^{57}\text{Co}$  rhodium matrix point-source with a nominal activity of 0.37 GBq was used. The absorber was prepared by grinding a sample (ca 1 mg) together with a thermoplastic resin, which was then shaped to a ca 1-mm sized cylinder under mild heating ( $< 120\text{ }^{\circ}\text{C}$ ). The absorber was attached on strip tape and positioned close to the point source. The spectrum (Fig. 5) was collected for 900 h at room-



**Fig. 4** Polarized single-crystal FTIR spectra of potassic-richterite in the O-H stretching region. E is the electric field vector



**Fig. 5** Transmission  $^{57}\text{Fe}$  Mössbauer spectrum of potassic-richterite. The fitted S1 and S2 subspectra correspond to  $^{64}\text{Fe}^{3+}$  and  $^{44}\text{Fe}^{3+}$ , respectively

temperature over 1024 channels, covering the velocity range  $-4.2$  to  $+4.2\text{ mm}^{-1}$ , and was calibrated against an  $\alpha\text{-Fe}$  foil. Spectrum analysis was done assuming Lorentzian line shapes

**Table 2** X-ray powder diffraction pattern for potassic-richterite

$d_{\text{obs}}$ (Å)	$d_{\text{calc}}$ (Å)	$I_{\text{obs}}$ (%)	$I_{\text{calc}}$ (%)	$h$	$k$	$l$
9.05	9.02	5	44	0	2	0
<b>8.55</b>	<b>8.53</b>	<b>36</b>	<b>11</b>	<b>1</b>	<b>1</b>	<b>0</b>
4.51	4.51	8	37	0	4	0
4.27	4.27	4	3	2	2	0
3.40	3.40	20	100	1	3	1
<b>3.303</b>	<b>3.301</b>	<b>56</b>	<b>55</b>	<b>2</b>	<b>4</b>	<b>0</b>
<b>3.181</b>	<b>3.179</b>	<b>100</b>	<b>70</b>	<b>3</b>	<b>1</b>	<b>0</b>
2.970	2.971	15	31	2	2	1
<b>2.847</b>	<b>2.845</b>	<b>50</b>	<b>18</b>	<b>3</b>	<b>3</b>	<b>0</b>
2.756	2.756	18	32	-3	3	1
<b>2.714</b>	<b>2.714</b>	<b>37</b>	<b>98</b>	<b>1</b>	<b>5</b>	<b>1</b>
2.592	2.592	14	51	0	6	1
2.532	2.537	12	75	-2	0	2
2.407	2.406	12	7	3	5	0
2.353	2.352	23	40	-3	5	1
2.298	2.298	9	19	-1	7	1
2.173	2.174	25	34	2	6	1
2.035	2.036	7	7	3	5	1
1.927	1.926	22	8	5	1	0
1.667	1.668	27	28	4	6	1
1.650	1.650	10	6	4	8	0
1.617	1.617	12	5	1	11	0
1.521	1.520	5	7	1	9	2
1.513	1.512	6	24	-2	6	3
1.456	1.455	32	31	-6	6	1

The five strongest Bragg reflections are in bold

with the MossA program (Prescher et al. 2012). A relatively broad doublet (FWHM = 0.59) is dominant, with a centroid shift (CS) of 0.40(1) mm/s and quadrupole splitting (QS) of 0.65(1) mm/s, respectively, indicative of Fe<sup>3+</sup> at octahedrally coordinated cation sites. A small contribution, ca 4% of total absorption, from another doublet at CS = 0.09(2) and QS = 1.14(6) mm/s is attributed to Fe<sup>3+</sup> in tetrahedral coordination, in agreement with (Al + Si) ≤ 8 apfu in potassic-richterite. Any amount of Fe<sup>2+</sup>, if present, is estimated to be less than 2% of all Fe. The final  $\chi^2$  value of the spectrum fit was 1.45.

### X-ray diffraction data and structure refinement

X-ray powder diffraction data were recorded on a PANalytical X'Pert<sup>3</sup> Powder diffractometer equipped with an X'celerator silicon-strip detector and operated at 40 mA and 45 kV (CuK $\alpha$ -radiation,  $\lambda$  = 1.5406 Å). Peak positions were determined with the X'Pert HighScore Plus program and corrected against an external Si standard. Indexed  $d$  values and relative peak-heights above background are given in Table 2. The monoclinic unit-cell parameters, obtained by least-squares refinement of 22 reflections, are:  $a$  = 10.006(2) Å,  $b$  = 18.038(4) Å,  $c$  = 5.279(1) Å  $\beta$  = 104.5(3)° and  $V$  = 922.5(3) Å<sup>3</sup>.

A single-crystal X-ray study was done on a 420 × 80 × 90  $\mu$ m crystal using an Oxford Diffraction Xcalibur diffractometer, operating at 50 kV and 30 mA, with monochromatized MoK radiation and equipped with a CCD detector at 80 mm from the sample position. A combination of  $\omega/\phi$  scans, with a step scan of 1° and an exposure time of 25 s per frame at low theta angles and duplicating counting time at high theta angles, was used to maximize redundancy and data coverage. We collected high resolution data (up to 0.5 Å). Crystal data, including the refined unit-cell parameters, and experimental settings are summarized in Table 3.

The structure of potassic-richterite was refined starting from the atom coordinates of Oberti et al. (1992). Scattering curves for fully ionized chemical species were used at sites where chemical substitutions occur; neutral vs. ionized scattering curves were used at the  $T$  and anion sites [except O(3)]. Splitting of scattering at the sites  $M(4)$  and  $A$  was observed and added to the model as  $M(4')$ ,  $A(m)$  and  $A(2)$  sites, respectively. A maximum close to O(3) (ca. 0.8 Å) was also observed in the Fourier difference map and added to the model as the hydrogen position. The positions of  $A(m)$  and  $A(2)$  sites were fixed during refinement and the hydrogen isotropic-displacement parameter was restrained to the displacement parameter of the anions at O(3). Full-matrix least-squares refinement on  $F^2$  converged to  $R_{\text{obs}} = 3.64\%$  (1811 reflections with  $I_o > 2\sigma(I)$ ) and  $R_{\text{all}} = 6.23\%$  (2399 reflections). Refined atom coordinates and equivalent isotropic-displacement parameters are reported in Table 4. The anisotropic-displacement parameters are given in Table S1 (electronic supplementary). Selected interatomic distances and angles are given in

**Table 3** Crystal data and structure refinement for potassic-richterite

Temperature (K)	293(2)
Wavelength (Å)	0.71073
Crystal system	Monoclinic
Space group	$C2/m$
$a$ (Å)	9.9977(3)
$b$ (Å)	18.0409(4)
$c$ (Å)	5.2794(2)
$\beta$ (°)	104.465(4)
$V$ (Å <sup>3</sup> )	922.05(5)
$Z$	2
Calculated density (g·cm <sup>-3</sup> )	3.075
Absorption coefficient (mm <sup>-1</sup> )	1.040
$F(000)$	868
Crystal size (mm <sup>3</sup> )	0.42 × 0.09 × 0.08
Range of data collection (° $\theta$ )	3.99 to 45.62
Index ranges	$-18 \leq h \leq 18$ , $-35 \leq k \leq 36$ , $-7 \leq l \leq 6$
Reflections collected	19,458
Independent reflections	2399 [ $R_{\text{int}} = 0.0391$ ]
Completeness to theta = 25.00° (%)	99.4
Refinement method	Full-matrix least-squares on $F^2$
Data / restraints / parameters	2399 / 1 / 126
Goodness-of-fit on $F^2$	1.144
Final $R$ indices [ $I > 2\sigma(I)$ ]	$R1 = 0.0363$ , $wR2 = 0.0558$
$R$ indices (all data)	$R1 = 0.0623$ , $wR2 = 0.0631$
Largest diffraction peak and hole (e·Å <sup>-3</sup> )	0.521 and $-0.525$

Table 5. Site populations for potassic-richterite were derived from the unit-formula and the results of the structure refinement, and are given in Table 6.

On the basis of the observed mean bond lengths, and in agreement with what has been already observed by Oberti et al. (1993) for <sup>C</sup>Mn<sup>2+</sup>, the following (strong) Mn site preference is inferred:  $M(2) \gg M(1) > M(3)$ . Fe<sup>3+</sup> is also assumed to be ordered at the  $M(2)$  site, obtaining a site population of (Mg<sub>1.41</sub>Mn<sub>0.39</sub>Fe<sup>3+</sup><sub>0.20</sub>). This leaves 0.08 Mn<sup>2+</sup> to be assigned to  $M(1)$  and  $M(3)$  as well as 0.07 apfu at the  $M(4')$  site (see below). According to Table 6, the composition of the  $M(1)$  site is (Mg<sub>1.93</sub>Mn<sup>2+</sup><sub>0.07</sub>) and  $M(3)$  is (Mg<sub>0.97</sub>Mn<sup>2+</sup><sub>0.03</sub>), leaving some Mn<sup>2+</sup> to be allocated to  $M(4')$ .

The cations at the  $A$  sites are preferentially ordered. K plus some Na are ordered at  $A(m)$ ; the refined site-scattering at  $A(m)$  is 12.92 epfu, which corresponds to 0.61 K apfu and 0.14 Na apfu. The rest of Na is ordered at the  $A(2/m)$  site; the refined site-scattering at  $A(2/m)$  is 0.95 epfu, which accounts for 0.09 Na apfu. Regarding the  $A(2)$  subsite, the refined site scattering is 1.58 epfu, which accounts for 0.02 Pb apfu. Both K and Na contents at the  $A$  sites agree reasonably

**Table 4** Refined atom coordinates and equivalent isotropic displacement parameters

Atom	Site occupancy	$x/a$	$y/b$	$z/c$	$U_{eq}$
O(1)	O <sup>0</sup> 0.18(4) O <sup>−</sup> 0.82(4)	0.11078(9)	0.08613(4)	0.21789(19)	0.0077(2)
O(2)	O <sup>0</sup> 0.14(4) O <sup>−</sup> 0.82(4)	0.11899(9)	0.17008(5)	0.72515(19)	0.0088(2)
O(3)	O <sup>−</sup> 0.76(3) F <sup>−</sup> 0.24(3)	0.10599(15)	0	0.7149(3)	0.0100(4)
O(4)	O <sup>0</sup> 0.21(4) O <sup>−</sup> 0.79(4)	0.36222(10)	0.24756(5)	0.7947(2)	0.0124(2)
O(5)	O <sup>0</sup> 0.27(4) O <sup>−</sup> 0.73(4)	0.34366(10)	0.12997(5)	0.0920(2)	0.0109(2)
O(6)	O <sup>0</sup> 0.26(4) O <sup>−</sup> 0.74(4)	0.34035(10)	0.11635(5)	0.5911(2)	0.0109(2)
O(7)	O <sup>0</sup> 0.26(6) O <sup>−</sup> 0.74(6)	0.33358(14)	0	0.2945(3)	0.0118(3)
T(1)	Si <sup>0</sup> 0.62(3) Si <sup>4+</sup> 0.38(3)	0.27584(4)	0.084927(16)	0.29666(8)	0.00652(9)
T(2)	Si <sup>0</sup> 0.64(3) Si <sup>4+</sup> 0.36(3)	0.28522(4)	0.171242(17)	0.80204(8)	0.00675(9)
M(1)	Mg <sup>2+</sup> 0.960(3) Mn <sup>2+</sup> 0.040(3)	0	0.08917(3)	½	0.0077(2)
M(2)	Mg <sup>2+</sup> 0.692(3) Mn <sup>2+</sup> 0.308(3)	0	0.18073(2)	0	0.00865(16)
M(3)	Mg <sup>2+</sup> 0.973(4) Mn <sup>2+</sup> 0.027(4)	0	0	0	0.0074(3)
M(4)	Ca <sup>2+</sup> 0.51(3) Na <sup>+</sup> 0.42(7)	0	0.2774(3)	½	0.0135(5)
M(4')	Mn <sup>2+</sup> 0.06(2)	0	0.264(3)	½	0.015(3)
A(m)	K <sup>+</sup> 0.340(5)	0.0137	½	0.0347	0.0430(13)
A	Na <sup>+</sup> 0.087(14)	0	½	0	0.009
A2	Na <sup>+</sup> 0.072(7)	0	0.4669	0	0.043(8)
H	H 0.76(3)	0.181(6)	0.0000	0.755(10)	0.048(15)

well with the EPMA analysis, (K<sub>0.61</sub>Na<sub>0.30</sub>Pb<sub>0.02</sub>). The chemical analyses indicate nearly full occupancy of the A sites (7% vacancy), and the above site assignment from structure refinement agrees well with this value. The nearly complete occupancy also indicated in the FTIR spectrum, where a weaker band is observed at 3672 cm<sup>−1</sup>, compatible with a tremolite-like environment (i.e. <sup>Δ</sup>□-MgMgMg-SiSi). The existence of A(2) could be related to the presence of “small” divalent cations at the B position, but also to the ordering of Pb at the A-sites observed for joesmithite (Moore 1968, 1969) and plumboan pargasite (Hålenius and Bosi 2012), where the Pb atoms orders at the A sites and shifts away from the inversion center along the diad, in the case of joesmithite leading to a decrease in symmetry from C2/m to P2/a (Moore et al. 1993). There is a significant occupancy at the B-group by Mn<sup>2+</sup>, which enters the M(4') site displaced towards the strip of octahedra relative to the M(4) site, as observed by Oberti et al. (1993) in some samples of Mn-bearing richterite. The worst agreement between observed (refined) and calculated

(assigned) site populations is observed for the B sites and A sites (Table 6), where differences of up to 7–8% are thought to be due to NaMn<sup>2+</sup><sub>−1</sub> variability between single analytical points (Table 1). Such a zonation is visible in Fig. 2.

The observed <T(1)-O> distance is somewhat longer than expected from the small amounts of Al<sup>3+</sup> measured by EPMA analysis. The equation for calculating <sup>T(1)</sup>Al from <T(1)-O> distance proposed by Oberti et al. (2007a) gives 0.13 apfu vs. the analysed amount of 0.04 apfu. Similar values of <T(1)-O> were reported for potassic-richterite by Oberti et al. (1992), although some of their crystals had slightly lower Al content (1.622 Å and 0.05 apfu Al in T(1)). Using the observed <T(2)-O> distance and the predictive equation of Oberti et al. (2007a), up to 0.19 apfu of Al in T(2) is indicated, which is not in agreement with the EPMA analysis. On the other hand, the EPMA analysis gave no Ti and a deficiency of Al + Si up to 0.04 apfu for certain points. If a large cation like Fe<sup>3+</sup> is to occupy a T-site in richterite, following Oberti et al. (1992), it occurs at the T(2) site. This would certainly enlarge

**Table 5** Selected interatomic distances and bond angles in potassic-rich richterite

Interatomic bond	Distance (Å)	Interatomic bond	Distance (Å)
<i>T</i> (1)–O(1)	1.598(1)	<i>T</i> (2)–O(2)	1.609(1)
<i>T</i> (1)–O(5)	1.629(1)	<i>T</i> (2)–O(4)	1.583(1)
<i>T</i> (1)–O(6)	1.629(1)	<i>T</i> (2)–O(5)	1.683(1)
<i>T</i> (1)–O(7)	1.638(1)	<i>T</i> (2)–O(6)	1.671(1)
< <i>T</i> (1)–O>	1.624	< <i>T</i> (2)–O>	1.636
<i>M</i> (1)–O(1) × 2	2.067 (1)	<i>M</i> (2)–O(1) × 2	2.197(1)
<i>M</i> (1)–O(2) × 2	2.062(1)	<i>M</i> (2)–O(2) × 2	2.102(1)
<i>M</i> (1)–O(3) × 2	2.097(1)	<i>M</i> (2)–O(4) × 2	2.000(1)
< <i>M</i> (1)–O>	2.075	< <i>M</i> (2)–O>	2.100
<i>M</i> (3)–O(1) × 4	2.080(1)	<i>M</i> (4)–O(2) × 2	2.419(4)
<i>M</i> (3)–O(3) × 2	2.046(1)	<i>M</i> (4)–O(4) × 2	2.364 (1)
< <i>M</i> (3)–O>	2.069	<i>M</i> (4)–O(5) × 2	2.866(2)
		<i>M</i> (4)–O(6) × 2	2.619(3)
<i>A</i> –O(5) × 4	2.951(1)	< <i>M</i> (4)–O>	2.567
<i>A</i> –O(6) × 4	3.145(1)		
<i>A</i> –O(7) × 2	2.545(1)	<i>M</i> (4′)–O(2) × 2	2.19 (5)
< <i>A</i> –O>	2.937	<i>M</i> (4′)–O(4) × 2	2.327(4)
		<i>M</i> (4′)–O(5) × 2	3.04(4)
<i>A</i> (2)–O(5) × 2	2.472(1)	<i>M</i> (4′)–O(6) × 2	2.84(5)
<i>A</i> (2)–O(6) × 2	2.783(1)	< <sup>[6]</sup> <i>M</i> (4′)–O>	2.567
<i>A</i> (2)–O(7) × 2	2.614(2)	< <sup>[8]</sup> <i>M</i> (4′)–O>	2.600
< <i>A</i> (2)–O>	2.623		
		Interatomic bond	Angle (°)
<i>A</i> ( <i>m</i> )–O(5) × 2	2.956(1)	<i>T</i> (1)–O(5)– <i>T</i> (2)	136.20(7)
<i>A</i> ( <i>m</i> )–O(5) × 2	2.908(1)	<i>T</i> (1)–O(6)– <i>T</i> (2)	136.01(7)
<i>A</i> ( <i>m</i> )–O(6) × 2	3.000(1)	<i>T</i> (1)–O(7)– <i>T</i> (1)	138.5(1)
<i>A</i> ( <i>m</i> )–O(7)	2.521(2)	O(5)–O(6)–O(5)	169.27(6)
<i>A</i> ( <i>m</i> )–O(6) × 2	3.297(1)	O(6)–O(7)–O(6)	107.10(8)
<i>A</i> ( <i>m</i> )–O(7)	2.584(2)		
< <i>A</i> ( <i>m</i> )–O>	2.943		
O(3)–H	0.73(6)		

**Table 6** Site populations for potassic-rich richterite

Site	Site population (apfu)	ss (epfu)		mbl (Å)	
		refined	calculated	refined	calculated
<i>T</i> (1)	3.96 Si <sup>4+</sup> + 0.04 Al <sup>3+</sup>			1.624	1.623
<i>T</i> (2)	3.98 Si <sup>4+</sup> + 0.02 Fe <sup>3+</sup>			1.636	
<i>M</i> (1)	1.93 Mg <sup>2+</sup> + 0.07 Mn <sup>2+</sup>	25.04	24.91	2.075	2.073
<i>M</i> (2)	1.41 Mg <sup>2+</sup> + 0.39 Mn <sup>2+</sup> + 0.20 Fe <sup>3+</sup>	32.00	31.87	2.100	2.098
<i>M</i> (3)	0.97 Mg <sup>2+</sup> + 0.03 Mn <sup>2+</sup>	12.35	12.39	2.069	2.075
Σ C cations		69.40	69.17		
B cations	1.14 Na + 0.07 Mn <sup>2+</sup> + 0.79 Ca <sup>2+</sup>	32.64	30.09		
A cations	0.61 K <sup>+</sup> + 0.30 Na <sup>+</sup> + 0.02 Pb <sup>2+</sup>	15.45	16.53		
W anions	1.82 OH + 0.18 F	16.48	16.18		

There is a good agreement between the refined values of site-scattering (ss) - given in electrons per formula unit (epfu) - and mean bond-lengths (mbl, in Å), and those calculated based on the proposed site populations

the size of the *T*(2) tetrahedron, even for a very low amount, justifying the observed <*T*(2)–O> distance (1.636 Å), that is slightly larger than observed for the <sup>IV</sup>Ti-free richterite samples studied by Oberti et al. (1992). Although the amount is so low that it is within the error of the analytical methods, the fitting of a weak <sup>IV</sup>Fe<sup>3+</sup> doublet to the Mössbauer spectrum seems to agree well with this hypothesis.

## Petrogenesis

Potassic-rich richterite is a rare member of the amphibole supergroup but occurs over a wide range of geological environments and *P*–*T* conditions. Common features are silica undersaturation and peralkaline bulk compositions of the rock system, although a few exceptions to this are found. The high-*T* and high-*P* stability, relevant for upper mantle conditions, has been studied in some detail (e.g. Konzett et al. 1997; Trønnes 2002). Overall, the thermal stability increases as a function of increasing *P*, and K-rich compositions tend to persist to higher *P* and *T*. There is also experimental evidence that an increase of the F/OH ratio stabilizes potassic-(fluoro)-richterite to higher *T* (Foley 1991). More generally, successful synthesis of near end-member potassic-rich richterite has been done from 700 °C and at standard conditions of *P* = 1–2 kbar (H<sub>2</sub>O), with good yields and few microstructural defects (e.g. Zimmermann et al. 1997; Hawthorne et al. 1997). However, a small excess of Ca (i.e. a tremolite component) is not uncommon in synthetic samples (e.g., Melzer et al. 2000). Data on the stability and phase relations at lower *T* and *P* are apparently missing. The assemblage with potassic-rich richterite from a blueschist facies terrain (Mottana and Griffin 1986) suggests that formation is possible at lower *T*, < 450 °C.

It is probable that different sources of key elements contributed to the eventual formation of potassic-



richterite. The marble-ore precursors contain the necessary Ca, Mg and Si, with additional Mn, Fe and Pb, in the Långban-type deposits (Magnusson 1930; Holtstam and Mansfeld 2001). K, Na and to some extent Si, have likely been added from fluids via metasomatic alteration of the felsic volcanic beds. At Pajsberg, potassic-richterite most probably crystallized during the main skarn-forming process, synchronous with Svecokarelian ~1.85 Ga peak regional metamorphism, inferred at temperatures of ca 700 °C and pressures of 3–6 kb in this part of Bergslagen (Stephens et al. 2009). These conditions are similar to the estimates of Grew et al. (1994),  $P \leq 4$  kbar,  $T \geq 500$  °C, and of Christy and Gatedal (2005),  $P = 3$  kbar,  $T > 600$  °C, for skarns in the Långban deposit, 13 km N of Pajsberg. It is possible that the mineralization also was affected by contact metamorphism from a small body of metabasite in the immediate vicinity of the Pajsberg field, appearing along the contact between the metavolcanic country rock and the metacarbonate host (Björck 1986). As estimated from the association of potassic-richterite with tephroite and  $Mn^{3+}$ -bearing jacobsonite, oxygen fugacity during mineral formation was above the magnetite stability field but well below that of bixbyite (Brunsitsyn 2007). As noted, the type specimen is free of Ti, in contrast to most samples of potassic-richterite from mantle-derived rocks. Due to geometrical constraints, Ti incorporation at the T(2) site is believed to be favoured by dominance of K at the A site (Oberti et al. 2007b).

Metamorphic amphiboles with  $[A]K > [A]Na$  are generally rare, and in a collection of more than 250 analyses of sodium-calcium amphiboles from metamorphic assemblages, only a handful were observed to contain  $>0.3$  K atoms pfu (Schumacher 2007). The K/Na ratios of richterite essentially depends on the activity ratio of these elements in the mineral-forming fluid system (Zimmermann et al. 1997; Safonov and Butvina 2016). For K-rich amphibole compositions to form, it is probably critical that  $SiO_2$  and  $Al_2O_3$  are low, so that K is not bound up in other silicate minerals such as K-feldspar or trioctahedral mica.

The paragenetic relations and premetamorphic precursors to potassic-richterite in the present occurrence cannot be determined with certainty. It is possible that the mineral formed from direct reactions between phlogopite, dolomite and quartz, also involving a Na-rich fluid; however, the very low Al content of the amphibole produced remains to be explained. Another scenario is that an alkali-rich hydrothermal fluid has reacted with pre-existing tephroite and dolomite during the skarn-forming process, giving potassic-richterite and jacobsonite.

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