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Pink zoisite from the Aar Massif, Switzerland

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ABSTRACT. Veins in the Central Aar granite consisting of zoisite, plagioclase, albite, alkali feldspar, muscovite, tremolite, quartz, and calcite are described. The zoisite is a thulite with up to 0.20 wt % MnO. It is argued that the zoisite is derived from granite plagioclases and has recrystallized in veins during alpine greenschist metamorphism. By later increasing X_{CO_2} or decreasing temperature zoisite decomposed to calcite, plagioclase, and muscovite.

Thulites from different localities were investigated by X-ray and by electron microprobe analysis. Both orthorhombic and monoclinic members of the epidote group having low MnO contents have been described as thulites. It is suggested that low MnO values in the range 0.05 to 0.9 wt % cause the pink colour of thulites, regardless of symmetry.

DURING field-work in a new tunnel of the KWO hydroelectric power plant near the Grimsel Pass in the Central Aar Massif (Switzerland), pink zoisite (thulite) has been found in a fine-grained vein coexisting with plagioclase (An_{17-24}), alkali feldspar, albite, quartz, muscovite, tremolite, and calcite. The host rock is a coarse-grained, light-green meta-granite (Central Aar granite, Labhart, 1977). The granite shows a polyphase alpine deformation beginning with a development of the dominant foliation striking north-east ($= S_1$), followed by different stages of deformation during which several new mineral phases have been formed. Veins consisting of milky quartz or quartz and carbonate are characteristic for the S_3-phase corresponding to an east-west striking fault system. The thulite-bearing veins parallel these quartz veins and neither show the older S_1 (south-west-north-east) schistosity suggesting a contemporaneous formation of the thulite-bearing and the quartz-carbonate veins after the S_1 event.

Microscopic description and chemical data†

The pink veins are 1 to 5 cm thick and often zoned with a layer of light-green epidote and quartz next to the granite. Mineral assemblages vary from vein to vein, but zoisite and carbonate are always present. The following assemblages were observed in different veins:

Sample Th 12 a: Zo + Cc + Plg + Mu + Ab + Qtz ± Tr
Th 12 b: Zo + Cc + Mu + Kfs + Qtz
Th 12 c: Zo + Cc + Kfs + Qtz + Tr
Th 12 d: Zo + Cc + Plg + Qtz

Zoisite. Zoisite shows a very uneven distribution and occurs as small prisms up to 0.5 mm in size, as very fine-grained, xenomorphic, pigmented masses, or as skeletal grains with muscovite and carbonate forming along the cleavage planes (fig. 1).

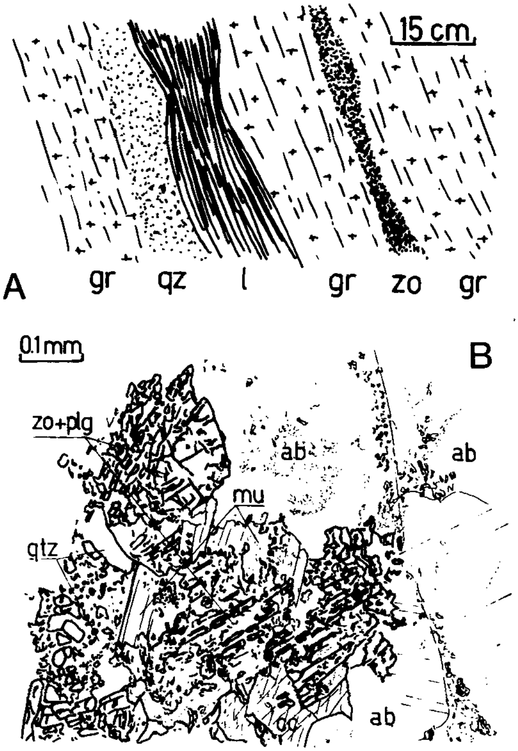

FIG. 1. (*a*) Zoisite (zo) vein in granite (gr); qz: quartz vein, l: lamprophyre. (*b*) Skeletal zoisite with plagioclase, muscovite, calcite, quartz, and albite.

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† The chemical data given in this paper were collected with an ARL-SEMQ microprobe using a ZAF-program for matrix corrections.

TABLE I. *Electron microprobe analyses of zoisites from the Aar Massif*

	$\bar{X}$	σ	Structural formula	
SiO_2	39.54	0.3	5.975	Si
Al_2O_3	33.08	0.5	0.025 5.868	Al^{IV} Al^{VI}
Fe_2O_3	0.61	0.3	0.069	Fe^3
MnO	0.07	0.08	0.009	Mn
MgO	0.32	0.12	0.072	Mg
CaO	24.84	0.09	4.021	Ca
Na_2O	0.03	0.04	0.009	Na
K_2O	—			
Total	98.43			

Zoisite, Aar Massif, mean ($\bar{X}$) and standard deviation (σ) of eleven analyses. Number of ions calculated on anhydrous basis of 25(O). FeO values lie between 0.19 and 1.09 wt%.

MnO values lie between 0.0 and 0.20 wt%.

The orientation of the optic plane is perpendicular to the (100) cleavage indicating that the material is β-zoisite. γ-α is estimated to be between 0.005 and 0.007. The average of eleven electron-probe analyses is given in Table I. Some clinozoisite grains with anomalous blue interference colours were observed together with zoisite.

Alkali feldspar. Alkali feldspar often shows a pigmented core and an inclusion-free border. In contrast to the alkali feldspar in the granite, the vein alkali feldspars are not perthitic. They have a mean Or-content of 92.3 mole% to 97.6 mole% and an An content of about 0.1 mole%. No difference in composition was measured between pigmented core and edge.

Plagioclase. The plagioclase was only found by microprobe analysis as small grains in and around xenomorphic zoisite masses. It is an oligoclase with a mean An content of 19.0 mole% (7 analyses). No zonation has been observed.

Albite. Albite shows inclusion-free grains that sometimes are twinned on (010). Its mean An content is 1.4 mole% and An contents lie in the range of 0.3 to 6.7 mole% (12 analyses).

Muscovite. Muscovite occurs in small flakes, sometimes along cracks in zoisite. Its growth seems to have been at the expense of zoisite. A representative analysis gave the formula* $K_{2.02}(Na,Ca)_{0.09}Mg_{0.50}Fe^{3+}_{0.10}Al^{VI}_{3.40}[Si_{6.3}Al_{1.7}O_{20}](OH)_4$.

All eight analysed muscovites have between 2 and 4 wt% MgO and Na+Ca below 0.1 (per formula unit).

Calcite. The carbonate forms xenomorphic interstitial grains of different size mostly together with zoisite and muscovite. All calcites analysed were nearly pure with about 98 mole% $CaCO_3$.

Tremolite. Amphibole occurs either as isolated elongated prisms or as thin radiating needles included in quartz or alkali feldspar. A representative analysis gave the formula* $Ca_{2.0}(Na,K)_{0.07}Mg_{4.80}(Fe^{3+},Al^{VI})_{0.30}[Si_{7.8}Al_{0.2}O_{22}](OH)_2$.

Genetic considerations

Based on field observations, the thulite vein formed during alpine metamorphism. Epidote minerals formed from plagioclase are found very frequently, either as small inclusions in plagioclase or as veins along cleavage planes (Stalder, 1964; Steck, 1968). Decomposition is enhanced in zones of strong tectonic deformation (Stalder, 1964). It is suggested that the thulites together with albite, muscovite, and calcite have been formed by such decomposition and recrystallized together with quartz, albite, and alkali feldspar in veins along slip-fault zones. The oligoclase found with thulite is considered as a late reaction product of the thulite and not as a pre-alpine relict of primary plagioclase.

All the present mineral phases can be represented in the iron-free system MgO-CaO-K_2O-Na_2O-Al_2O_3-SiO_2-H_2O-CO_2, where the two following reactions are likely to have occurred

$$2\,Zo + 3\,Kfs + 2\,H_2O + 4\,CO_2 \rightleftharpoons 3\,Mu + 4\,Cc + 6\,Qtz \quad [1]$$

$$Zo + Ab_{ss} + CO_2 \rightleftharpoons An_{ss} + Cc + H_2O \quad [2]$$

The assemblage Mu + Ksp + Zo + Plg + Qtz + Cc + V in our samples corresponds to the invariant point formed at the intersection of reaction [1] and [2] in a T-X_{CO_2} diagram (Hewitt, 1973). The position of that invariant point would give an indication of the conditions of formation of the above assemblage. Using the data of Storre and Nitsch (1972) and Hewitt (1973) the point is estimated to lie at ~ 380 °C at very low X_{CO_2} below 0.05 for a $P_f \sim 2$ kbar and a 20% anorthite component.

The reactions [1] and [2] take place either under decreasing temperature or increasing X_{CO_2}. An upper-temperature estimate may be obtained by the determination of triclinity on a concentrate of alkali feldspar of sample Th 12c by the powder diffraction method (Guinier-de Wolff camera, Fe-$K\alpha$ radiation), which gave a value of 0.7–0.8. According to Bernotat and Bambauer (1979) this would mean that a temperature of about 450 °C

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* Calculated on anhydrous basis of 22(O).

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TABLE II. *Thulite samples examined*

Sample no.	Locality	Colour*	Structure	Fe_p^{3+}†	Occurrence, assemblage	Reference
Th 1	P. Mezzogiorno Tessin, Switz.	Moderate pink 5R 7/4	mcl	0.03	+qtz+epidote (green)	Niggli *et al.* (p. 456, 1940)
Th 2	Lexviken, Trondheim, Norway	Moderate pink 5R 7/4	orh		+qtz+epidote	
Th 3	Zillertal, Austria	Moderate red 5R 5/4	mcl	0.30	Crystals up to 1 cm on cleavage plane	
Th 5	North Carolina, Yancey Co.	Moderate red 5R 5/4	mcl	0.01	Massive masses of crystals (to 2 mm) in albite pegmatite	(Schaller and Glass, 1942)
Th 6	Lexviken, Trondheim, Norway	Moderate pink to light red 5R 6/4	orh		+qtz+piemontite+amphibole. Very similar to Th 14 (wrong locality??)	
Th. 8	Telemarken, Norway	Light red 5R 6/6	orh		+qtz+idocrase	
Th 9	Praegraten, Austria	Moderate pink 5R 7/4	mcl	0.33	Small crystals on gneiss cleavage plane	
Th 10	Mte. Cadino, Adamello, Italy	Moderate pink 5R 7/4	mcl	0.03	In veins at granite-limestone contact, alteration product of plg, hbl	Bianchi and Dal Piaz (1937)
Th 11	Ravinella di Sotto, Italy	Pale to greyish red purple 5RP 5/2	mcl	0.25	Alteration product of plagioclase	Bertolani (1964) Abrecht *et al.* (1978)
Th 12 a–e	Grimsel Pass, Switzerland	Greyish pink 5R 8/2	orh		Alteration product of plagioclase in veins	This paper
Th 13	Arendal, Norway	Light red 5R 6/6	orh		+qtz (see no. Th 6) } ? same locality	
Th 14	Telemarken, Norway	Light red 5R 6/6	orh		+qtz	

* According to Rock colour chart GSA, 1951. † Fe_p^{3+}: Formula written as $Ca_2Al_{3-p}Fe_p^{3+}[Si_3O_{12}]OH$.

was not exceeded during alpine crystallization of the vein alkali feldspar.

Mg for the formation of tremolite probably comes from decomposition of granite biotite and/or chlorite, as these are the only Mg-bearing minerals in the Aar granite. Amphiboles are absent.

Comparison with other thulites

Zoisites with pink to purplish colour are described in the literature but usually without further specifications. Samples from nine different localities, detailed in Table II, have been studied by X-ray and electron microprobe. Samples were kindly provided by Professor Dr H. A. Stalder (Berne), Professor Dr C. Guillemin (Paris), and the collection of the Mineralogical and Petrographic Institute of the University of Berne. All were labelled as thulite. Thulite is a manganese-bearing zoisite according to different authors (Schaller and Glass, 1942; Deer *et al.*, 1962). The colour of the samples varies in its intensity between pale pink and purplish.

X-ray studies. All samples were studied by X-ray powder diffraction with a Guinier-de Wolff camera (Fe-$K\alpha$ radiation). A distinction between orthorhombic and monoclinic zoisite and clinozoisite/epidote, respectively, is easy by this method (Seki, 1959). These studies showed that six of nine 'thulites' had monoclinic patterns. Only the samples from Telemarken (Th 8, 13, 14), Lexviken (Th 2 and 6), and the Aar massif (Th 12) were orthozoisites. Pink clinozoisite from the Koralm (Austria) was reported by Meixner (1954). The name thulite has so far been used for pink zoisites. To eliminate confusion with piemontite the name Mn-epidote cannot be used for low-Mn^{2+} epidotes, so a distinction should be made according to the structure: monoclinic or orthorhombic thulite.

From the monoclinic samples the (020) reflection was measured to determine the approximate value of Fe^{3+} according to Myer (1965), the results are listed in Table II.

Additionally, from the zoisites Th 12a and Th 6 the cell parameters were determined by powder diffraction (Table III). The X-ray reflections were indexed according to Borg and Smith (1969) and the unit-cell calculations were made on the basis of the LATCON program (Schwarzenbach, ETH Zürich). The cell refinements were based on 55 and 43 peaks for sample Th 12a and Th 6, respectively.

TABLE III. *Unit cell parameters of zoisites*

p*	*a*	*b*	*c*	V(Å^3)	Ref.
0.05	16.205(2)	5.551(1)	10.035(1)	902.6(2)	1
0.07	16.199(4)	5.551(1)	10.033(2)	902.0(6)	2
0.07	16.207(3)	5.556(1)	10.036(2)	903.7(4)	2
0.09	16.212(8)	5.559(6)	10.036(4)	904.5	3
0.25†	16.177(6)	5.575(2)	10.045(3)	905.8(5)	3

* P: Formula written as $Ca_2Al_{3\text{-}p}Fe_p^{3+}[Si_3O_{12}]OH$.
† Synthetic Mn^{3+} zoisite.

References:
1. Myer (1966).
2. This paper.
3. Anastasiou and Langer (1977).

As would be expected the data given in Table III show an increase of the unit cell volume of zoisites with increasing Fe^{3+} substitution for Al^{VI}.

Chemical data and origin of pink colour. Different workers agree that manganese produces the pink colour of thulites but they disagree as to whether the manganese is di- or trivalent. Anastasiou and Langer (1977) synthesized a Mn^{3+}-bearing orthozoisite, which they called thulite. Strens (1966) reports a pink zoisite with 0.2 % of Mn_2O_3. Ghose and Tsang (1971), however, studied pink zoisite from Fanny Gouge Mine (North Carolina) with about 0.1 % Mn^{2+} by the electron paramagnetic resonance technique. Schaller and Glass (1942) suggest that small quantities of manganic manganese together with low Fe^{3+} contents cause the pink colour, a view that is supported by Lehmann (1978), who states that very pale-pink colours are produced when there is no other transition metal ion than Mn^{2+}.

This explanation seems favourable for our pale-pink thulite Th 12, which indeed has a very low iron content as well as the pink epidote from North Carolina (Th 5, Table IV) with FeO between 0 and 0.10 wt %. The zoisite from Lexviken (Th 6, Table IV) with dark-pink to purplish colour has higher FeO-values of up to 4.35 wt % (mean value: 1.65) and slightly higher MnO values between 0.20 and 0.86 wt %.

Fe_p^{3+} contents of pink epidotes determined by X-ray diffraction were below 0.33 (when $Ca_2Al_{3\text{-}p}Fe_p^{3+}Si_3O_{12}OH$) which corresponds to about 1.2 wt % Fe_2O_3. Epidotes with similar MnO-values (up to 0.4 wt %) but high Fe^{3+} (around 10 wt % Fe_2O_3) generally have green colours.

Highest iron contents in pink zoisite/clinozoisite were detected in the sample Th 6 with 1.84 wt % Fe_2O_3. An analysis in Deer *et al.* (1963) gives 0.47 wt % MnO and 0.90 wt % Fe_2O_3. Schaller and Glass (1942) report values of 1.62 wt % MnO_2 for thulites from different localities.

Sample Th 12c and 12e were separated by heavy liquids. Because of the very intimate intergrowths of zoisite with quartz, muscovite, and/or feldspar only concentrates of about 80-90 % purity were obtained. For Th 12c 0.1 wt % MnO and for Th 12e 0.05 wt % MnO were measured by atomic absorption (values corrected for 100 % zoisite). For sample Th 6 a MnO-content of 0.6 wt % was obtained for a concentrate of about 95 % purity.

Calculating the manganese of the analyses as Mn^{3+} together with Fe^{3+} as substitution for Al^{VI} in octahedral positions we obtain no clear relationship between Al^{VI} and Mn^{3+} in any of the samples. Calculating manganese as divalent, we obtain for the Th 5 and 6 samples a linear relationship between Mn^{2+} and Ca^{2+}. There is also a clear linearity of the relation Al^{VI}-Fe^{3+} (fig. 2) suggesting that the Mn substitutes in the seven- or eight-coordinated Ca sites and that only Fe^{3+} and Al substitute in the octahedral position. This

TABLE IV. *Chemical analyses of thulites*

Th 6	$\bar{X}$	σ	Structural formula	
SiO_2	38.87	0.20	5.902	Si
Al_2O_3	32.30	1.02	0.098 5.683	Al^{IV} Al^{VI}
Fe_3O_3	1.84	1.33	0.210	Fe^{3+}
MnO	0.48	0.19	0.062	Mn
MgO	0.64	0.06	0.145	Mg
CaO	24.45	0.26	3.977	Ca
Na_2O	0.17	0.06	0.050	Na
K_2O	—	—		
TiO_2	0.01			
Total	98.76			
Th 5				
SiO_2	38.82	0.24	5.943	Si
Al_2O_3	33.06	0.25	0.057 5.909	Al^{IV} Al^{VI}
Fe_2O_3	0.10	—		
MnO	0.31	0.20	0.040	Mn
MgO	0.39	0.05	0.089	Mg
CaO	24.61	0.25	4.036	Ca
Na_2O	—			
K_2O	—			
TiO_2	—			
Total	97.20			

Th. 6. Thulite, orh, Lexviken. Mean ($\bar{X}$) and standard deviation (σ) of nine analyses. Number of ions calculated on anhydrous basis of 25(O).

Th. 5. Thulite, mcl, North Carolina. Mean ($\bar{X}$) and standard deviation (σ) of eight analyses. Number of ions calculated on anhydrous basis of 25(O).

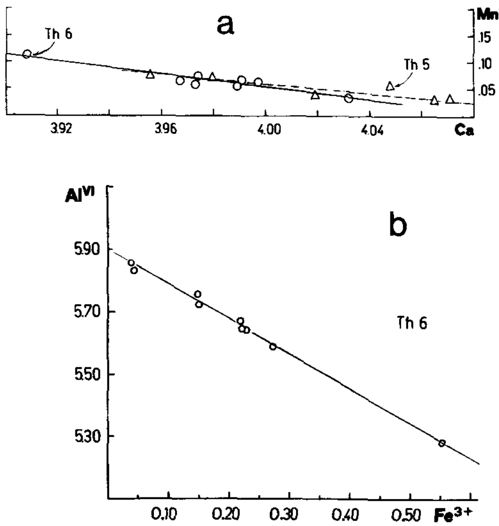

FIG. 2. (*a*) Plot of divalent Mn^{2+} and Ca of zoisites (sample Th 6) and clinozoisites (sample Th 5). Calculations on anhydrous basis of 25(O). (*b*) Proportions of octahedrally coordinated aluminium and ferric iron in thulites of sample Th 6. Calculations on anhydrous basis of 25(O).

supports the assumption of Mn as being in the divalent state in thulites. Fe^{3+} should be present only in small quantities for the pink colour.

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