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Davreuxite: a reinvestigation

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Abstract

Davreuxite from Ottré, Belgium, occurs as fibrous masses in quartz veins intimately associated with pyrophyllite and kaolinite. These quartz veins cross-cut the low-grade metamorphic manganese-rich rocks of Ordovician age of the Stavelot Massif, and also contain andalusite, ottrelite or chloritoid, sudoite, hematite, pyrophyllite, chlorite. Davreuxite is transparent, optically biaxial, negative; $\alpha = 1.660(5)$, $\beta = 1.684(2)$, $\gamma = 1.690(2)$; $2V_\alpha = 48(5)°$, $2V_\alpha$ (calc.) $= 52.5°$; $Z \parallel b$; $X \approx \perp (100)$. The mineral is monoclinic, space group $P2_1/m$, with $a = 9.550(2)$, $b = 5.767(1)$, $c = 12.077(2)$Å, $\beta = 108°1'(1')$; the ten most intense lines of the powder pattern are (d, I, hkl): 8.51(30)($10\bar{1}$); 5.719(35)($10\bar{2}$); 4.290(40)(102); 3.822(30)(003); 3.511(100)($20\bar{3}$); 3.179(30)($30\bar{1}$); 3.103(45)($30\bar{2}$); 2.870(60)(004); 2.840(35)($30\bar{3}$); 2.130(30)($40\bar{4}$). D(meas.) = 3.30 − 3.38; D(calc.) = 3.34 g/cm^3.

The ideal formula, derived from new microprobe analyses and from recent crystal structural data is $MnAl_6Si_4O_{17}(OH)_2$. The infrared spectrum is given. A comparison with davreuxite from Recht is also presented.

Historical background

Davreuxite was originally discovered between 1840 and 1850 by Dumont (1852) who simply called it "trémolite fibreuse" or "asbeste d'Ottré" and only mentioned its occurrence near the village of Ottré, in the quartz veins cross-cutting the manganiferous slates of the Cambro-Ordovician Massif of Stavelot, Belgium. Largely by comparing the specimens of A. Dumont with others collected in Salmchâteau and taking into account a wet chemical analysis, de Koninck (1878) obtained the composition $(Mn_{0.75}Mg_{0.25})Al_6Si_6\,O_{20}(OH)_4$ and named it davreuxite in honor of Charles Davreux (1800–1863), Belgian pharmacist and natural scientist, professor of mineralogy at the University of Liège, Belgium.

After an investigation of the optical properties Lacroix (1886) concluded that davreuxite was a "mica hydraté en lames fortement étirées". This conclusion seems to have introduced ambiguities, because Buttgenbach (1947) classified davreuxite in the mica group and Strunz (1970) considered it as a manganiferous vermiculite. Fransolet and Bourguignon (1976) therefore undertook to restudy the mineralogical properties of davreuxite and they proved that it is a valid species. These authors had difficulty obtaining pure samples for chemical analysis, as had de Koninck (1878). However, they proposed the ideal chemical formula $Mn_2Al_{12}Si_7O_{31}(OH)_6$. As the Al:Si ratio was thought to be inaccurate, they noted that a more accurate chemical analysis was necessary.

The purpose of the present study is to provide a complete mineralogical description of davreuxite by refining the previous data of Fransolet and Bourguignon (1976). Our conclusions are in large part based on the electron microprobe analysis and crystal structural analysis carried out by Jones et al. (1984).

Occurrence

Davreuxite has been found in the quartz veins outcropping in the southeastern border of the Cambro-Ordovician Stavelot Massif, Belgium. In this zone, quartz veins cross-cut the purplish-blue to purplish gray Salmian shales of Ordovician age; these are particularly rich in MnO, Fe_2O_3 and Al_2O_3 (Kramm, 1973). The rocks have been affected by low grade metamorphism of Hercynian age as recently corroborated by Schreyer (1975). The metapelites are characterized by varied mineralogical assemblages rich in Mn and Al; the minerals include

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Table 1. Davreuxite: Localities in the Stavelot Massif and associated minerals

Localities	Associated minerals	References
OTTRE (type locality)	Ottrelite, andalusite (pale blue) pyrophyllite, sudoite, kaolinite, rutile, dickite	Dumont (1852), de Rauw (1911), Malaise (1913), Fransolet (1978), Fransolet and Bourguignon (1976, 1978a, and 1978b), Kramm (1980).
SART-CLOSE (near Salmchâteau)	Chloritoid, hematite, chlorite	de Koninck (1878), Malaise (1913), Michot (1955).
VIELSALM	Andalusite, pyrophyllite	Malaise (1913).
REGNE	Andalusite (pale green), sudoite, pyrophyllite, hematite	Corin (1929), Fransolet and Bourguignon (1978b).
RECHT	Chloritoid, chlorite, hematite, pyrophyllite.	

spessartine, chloritoid, andalusite, and viridine (Kramm, 1973, 1976 and 1979). The low phengite component of the muscovites leads to a pressure of 1–2 kbar and, at these conditions, the presence of andalusite indicates temperatures of about 380°C (Kramm, 1973 and Fransolet et al., 1977).

According to Dumont (1852), de Koninck (1878), Malaise (1913), and Corin (1929), and our recent field observations, davreuxite occurs at five localities. The minerals associated with it in the quartz veins are listed in Table 1.

Physical properties

Davreuxite occurs as small bundles of extremely fine fibers which are cream-colored with yellow or very pale pink tinges. These bundles are enclosed in quartz, are up to several centimeters in length, and may be folded or imbricated. Davreuxite is brittle and breaks up into small blades following a relatively good cleavage {100} and cross-fractures {010}. The asbestos-like nature of davreuxite prevents an accurate determination of Mohs' hardness, which can only be tentatively estimated to be between 2 and 3.

Fibers alter to a dull, white product, which is a rather well-crystallized kaolinite (Fransolet and Bourguignon, 1978a). Complete pseudomorphs are observed under the polarizing microscope.

The presence of quartz between davreuxite fibers, the frequent alteration to kaolinite, and the common association with pyrophyllite cause density measurements to be unreliable, although Fransolet and Bourguignon (1976) reported D = 3.15(5). Our measurements using davreuxite from Ottré at 25°C in dilute Clerici solution yielded values between 3.12 and 3.30. On some coarser grains selected from optimally fresh samples, measurements using a torsion balance give a similar variation between 3.12 and 3.38. Thus the observed density of davreuxite is between 3.30 and 3.38.

As previously reported by de Koninck (1878), davreuxite is virtually insoluble in hot acids and a NaOH fusion is necessary to dissolve it. Davreuxite does not fluoresce in either short or long wavelength ultraviolet radiation.

Optical properties

Davreuxite is biaxially negative with $2V_\alpha$(obs.) = 48°(5°) as determined using Mallard's method. The refractive indices measured using sodium light are: $\alpha = 1.660(5)$, $\beta = 1.684(2)$ and $\gamma = 1.690(2)$. $2V_\alpha$(calc.) = (−)52.5°. The mean refractive index is 1.678 in good agreement with the computed index 1.696 from the Gladstone–Dale relationship (Mandarino, 1981). The value of the birefringence $\gamma - \alpha$ (0.030) is identical to that already observed by Lacroix (1886).

Under the microscope the mineral is colorless but a very faint yellowish tinge can be detected for the thicker grains. Crushed davreuxite yields many small laths with low birefringence which display the same optical orientation, i.e., virtually perpendicular to X, the acute bisectrix, as previously described by Lacroix (1886). Moreover, all grains have a parallel and positive extinction: $Z = b$ (the fiber axis). The optical investigation and the single-crystal measurements confirm the presence of a relatively good cleavage {100}, and show that the optic axial plane is parallel to b and virtually perpendicular to {100}. The fibrous habit prevents further refinements of the optical orientation.

Crystallography

On the basis of optical observations de Koninck (1878) assigned an orthorhombic symmetry to davreuxite. Fransolet and Bourguignon (1976) determined that it is monoclinic with $a = 9.57$, $b = 5.79$, c 12.88Å and $\beta = 116°$ using davreuxite from Ottré (specimen no. 8606-i.g. 5060, collection of the Institut royal des Sciences naturelles de Belgique, Bruxelles). After very long Weissenberg and precession exposures using an extremely small cleavage fragment (0.008 × 0.340 × 0.050 mm) selected from the same specimen, our recent measurements confirmed the lattice defined by Fransolet and Bourguignon (1976). However we have chosen to reindex on a reduced, less oblique cell with the new approximate dimensions: $a = 9.57$, $b = 5.79$, $c = 12.08$Å and $\beta = 108°$. The transformation formula from the former cell to the reduced one is $(\bar{1}00/0\bar{1}0/101)$. This very splinter served as a crystal of sufficient quality for the structure analysis (Jones et al., 1984). Weissenberg and precession data show that the space group is $P2_1/m$ or $P2_1$. The structural refinement (Jones et al., 1984) demonstrates that the space group is $P2_1/m$.

The X-ray powder diffraction pattern for davreuxite from Ottré, given in Table 2, were obtained with a diffractometer equipped with a monochromator, Fe$K\alpha$ radiation and an internal standard of $Pb(NO_3)_2$ (a = 7.8568Å). Indexing was performed taking into account the intensities of reflections as measured for the structure determination (Jones et al., 1984), and the intensities computed with the Program *Lazy Pulverix* (Yvon et al., 1977). Least-squares refinement using the program of Cox (1967) yielded the cell parameters for davreuxite from Ottré: a = 9.550(2), b = 5.767(1), c = 12.077(2)Å and β = 108°1'(1'). These values obtained from the powder data are somewhat different from those determined by Jones et al. (1984) on the only available single crystal, using a four-circle diffractometer. Without the chemical composition of this single crystal, this discrepancy cannot be explained.

Chemical composition

Because of the intimate intergrowth of davreuxite, quartz, and pyrophyllite, wet chemical analyses were inaccurate. de Koninck (1878) observed a noticeable contamination by quartz (Table 3, column 1). As Fransolet and Bourguignon (1976) detected quartz and pyrophyllite by X-ray diffraction of the selected material analyzed by J.-M. Speetjens (Table 3, column 2), they corrected for a content of 5 wt.% pyrophyllite and 10 wt.% quartz in the starting product (Table 3, column 3).

Electron microprobe analyses were therefore performed with a Cambridge Microscan MKV equipment (20 kV, and 30 nA) at six points on three different grains of davreuxite from Ottré. The synthetic compounds SiO_2, Al_2O_3,Fe_3O_4,Mn,MgO,$AlPO_4$(for P), $Na_2ZnSi_3O_8$(for Zn) and $CuSrSi_4O_{10}$(Cu) served as standards. The correction procedure outlined by Sweatmann and Long (1969) was used. A wavelength dispersive scan indicated the absence of any detectable element with atomic number greater than nine except for the ones reported here. The average of the six measurements for each element is presented in Table 3. The microprobe analysis shows that davreuxite from Ottré contains some phosphorus whose weight percent is approximately constant, and appreciable amounts of Cu and Zn. The weight percentages of these elements strongly vary from one point to the other in the same grain. We observe that the range for CuO variations is 0.47–1.17 wt.%, and for ZnO 0.27–0.68 wt.%.

The structure determination shows that there are 38 oxygen atoms per cell and that four hydrogen atoms are needed for charge balance (Jones et al., 1984). Consequently, the cation numbers were established on the basis of 36 cationic valences (Table 4) and the calculated water content is 2.83%. Using the refined cell parameters and with Z = 2, the calculated specific gravity is 3.34 g/cm^3, in excellent agreement with the highest values of the measured density. The deduced chemical formula is $(Mn_{0.82}Mg_{0.07}Cu_{0.06}Zn_{0.04})_{0.99}(Al_{5.86}Fe^{3+}_{0.09})_{5.95}(Si_{4.01}P_{0.03})_{4.04}O_{17}(OH)_2$ and the ideal chemical composition for davreuxite is $MnAl_6Si_4O_{17}(OH)_2$.

Table 2. X-ray powder diffraction data for davreuxite from Ottré

I/I₀*	d$_{obs.}$(Å)	hkl	d$_{calc.}$(Å)	I/I₀*	d$_{obs.}$(Å)	hkl	d$_{calc.}$(Å)
10	11.47	001	11.48	10	2.303	023	2.303
10	9.20	Pyrophyllite				221	2.305
30	8.51	$10\bar{1}$	8.52	5	2.287	$40\bar{3}$	2.288
5	7.16	Kaolinite		10	2.226	$22\bar{3}$	2.228
35	5.719	$10\bar{2}$	5.718	5	2.192	$41\bar{1}$	2.193
5	4.597	Pyrophyllite		5	2.144	204	2.145
10	4.538	200	4.541	30	2.130	$40\bar{4}$	2.130
40	4.290	102	4.290	10	2.079	105	2.079
10	4.260	$20\bar{2}$	4.260	5	2.034	024	2.034
5	4.062	$11\bar{2}$	4.060	5	2.022	$32\bar{3}$	2.023
10	3.996	$10\bar{3}$	3.998	25	1.999	$20\bar{6}$	1.999
30	3.822	003	3.828			$10\bar{6}$	2.002
10	3.569	210	3.567	10	1.914	006	1.914
100	3.511	$20\bar{3}$	3.511			223	1.915
5	3.342	Quartz		5	1.889	$21\bar{6}$	1.889
15	3.285	$11\bar{3}$	3.286	5	1.840	322	1.840
20	3.188	103	3.192	5	1.836	$42\bar{2}$	1.834
30	3.179	$30\bar{1}$	3.180	5	1.831	$42\bar{1}$	1.831
25	3.122	202	3.122	5	1.816	500	1.816
45	3.103	$30\bar{2}$	3.103	5	1.809	$31\bar{6}$	1.810
15	3.066	Pyrophyllite		5	1.714		
5	3.026	300	3.027	5	1.710		
60	2.870	004	2.871	5	1.676		
35	2.840	$30\bar{3}$	2.840	5	1.650		
5	2.731	$31\bar{2}$	2.733	5	1.642		
10	2.675	$11\bar{4}$	2.675	10	1.590		
5	2.618	121	2.618	10	1.584		
5	2.573	014	2.570	5	1.575		
5	2.547	$31\bar{3}$	2.548	5	1.561		
25	2.521	104	2.522	10	1.547		
25	2.505	$30\bar{4}$	2.506	10	1.532		
10	2.465	$22\bar{1}$	2.466				
5	2.410	$10\bar{5}$	2.411				
5	2.388	$22\bar{2}$	2.388				
5	2.365	$20\bar{5}$	2.365				

* Intensities are peak heights
Indexed on the basis of a monoclinic cell with
$\underline{a}$ = 9.550, $\underline{b}$ = 5.767, $\underline{c}$ = 12.077 Å and β = 108°1'
FeK_α radiation; $\underline{d}$ values corrected with a $Pb(NO_3)_2$ internal standard.

In order to obtain accurate water analyses, new attempts were made to purify fresh material collected by us in the Ottré quarry (magnetic and gravity separations, as well as sedimentation in water to eliminate the phyllosilicates) but these did not succeed completely. X-ray diffraction shows that the purest davreuxite samples contained a very low content of pyrophyllite, kaolinite and quartz as impurities. However, water determination by the Penfield procedure on this very material gave 2.96% H_2O, in rather good agreement with the calculated water content.

Infrared spectrum

The infrared spectrum of davreuxite (Fig. 1) was recorded with a Beckmann IR-12 spectrophotometer in the 200–4000 cm^{-1} region, on a sample of 4 mg diluted in 600 mg of KBr, prepared with the pressed pellets technique.

Although a full assignment of all the absorption peaks

Table 3. Wet chemical and electron microprobe analyses of davreuxite

	1	2	3	4		5		6
				a	b	a	b	
P_2O_5	-	-	-	0.35	0.031	-	-	-
SiO_2	39.31	42.83	34.42	37.82	4.009	37.45	3.979	37.84
TiO_2	-	-	-	-	-	tr.	-	-
Cr_2O_3	-	-	-	-	-	tr.	-	-
Al_2O_3	33.59	43.49	49.11	46.88	5.857	48.09	6.022	48.15
Fe_2O_3	tr.	1.05	1.23	1.10	0.088	-	-	-
FeO	-	-	-	-	-	1.29	0.115	-
MnO	5.25	8.85	10.33	9.08	0.815	9.14	0.822	11.17
MgO	1.10	0.48	0.56	0.44	0.070	0.45	0.071	-
ZnO	-	-	-	0.49	0.038	-	-	-
CuO	-	-	-	0.79	0.063	-	-	-
H_2O	4.19	3.98	4.35	[2.83]*	1.998	[2.82]*	2.000	2.84
Quartz	16.63	-	-	-		-		-
total	100.07	100.68	100.00	99.78		99.24		100.00

* Calculated wt% for H_2O

1. Davreuxite from Sart-Close, Salmchâteau (de Koninck, 1878)
2. Davreuxite from Ottré (Analyst: J.-M. Speetjens, *in* Fransolet and Bourguignon, 1976)
3. Recalculated analysis to 100%, after that 10 wt% quartz and 5 wt% pyrophyllite have been substracted from 2 (Fransolet and Bourguignon, 1976)
4. Davreuxite from Ottré
5. Davreuxite from Recht
6. Theoretical composition for $MnAl_6Si_4O_{17}(OH)_2$

a. Electron microprobe data (Analyst: K. Abraham)

b. Cation numbers on the basis of 36 cationic valences

visible in this complex infrared spectrum is not intended here, the spectrum indicates several features, confirmed by the results of the structure analysis (Jones et al., 1984): (1) the presence of OH groups whose stretching vibrations form an intense doublet at 3396 and 3478 cm^{-1} indicating by these frequencies the presence of hydrogen bonds; (2) determination of tetrahedral aluminium by the existence of the very intense band at 815 cm^{-1}; (3) occurrence of almost linear, strong Si–O–Al bonds in the structure, also indicated by another very intense band at 1200 cm^{-1}; (4) close spectral similarities with the infrared spectra of sillimanite (Tarte, 1969) permitted the prediction of the structural analogies between these minerals.

Comparison with davreuxite from Recht

More recently we have made an investigation of davreuxite from Recht (Table 1). Following the procedure used for the mineral from Ottré the calculation of cell parameters gives: a = 9.556(3), b = 5.763(2), c = 12.084(4)Å and β = 108°34'(1'). Compared with the single crystal data of Jones et al. (1984) an unexplained discrepancy is observed, as for the cell dimensions of Ottré davreuxite.

The chemical analysis was performed with a CAMEBAX microprobe, using synthetic andradite (Fe and Si), synthetic pyrope (Mg and Al) and Mn (Mn) as standards, and

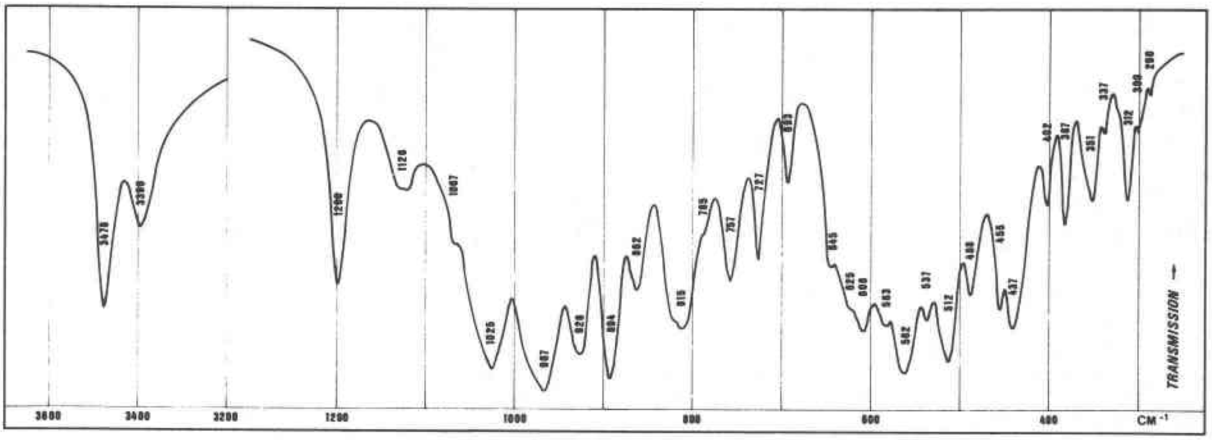

Fig. 1. Infrared spectrum of davreuxite from Ottré.

the same ZAF-correction used before. The average of the eight point-analyses is given in Table 3. Davreuxite from Recht does not have P, Cu and Zn, but traces of Ti and Cr were detected.

The molecular proportions confirm the ratio Si:Al:Mn = 4:3:1. The cation numbers on the basis of 36 cationic valences are given in Table 3. With a calculated water content of 2.82%, the refined cell parameters, and Z = 2, the calculated density is 3.33 g/cm^3 for a chemical formula $(Mn_{0.82}Fe_{0.12}Mg_{0.07})_{1.01}Al_{6.02}Si_{3.98}O_{17}(OH)_2$, in very good agreement with the ideal formula $MnAl_6Si_4O_{17}(OH)_2$.

For davreuxite from Ottré, Fe has been calculated as Fe_2O_3, whereas it is given as FeO in this analysis. This was done to optimize the stoichiometry, $R^{2+}:R^{3+} = 1:6$.

Compared with the results obtained for the Recht material, the presence of P, Cu and Zn in Ottré davreuxite is puzzling. The virtually constant weight content of P_2O_5 and the systematic absence of Ca in the latter constitute an argument to reject the hypothesis of the presence of apatite as a supplementary impurity. Moreover, the P^{5+} substitution on the tetrahedral sites has been recently demonstrated in the blue andalusite from Ottré, occurring in the same quartz vein (Prof. K. Langer, private comm.). For these reasons we suggest that a small amount of P^{5+} is incorporated on tetrahedral Si-sites of davreuxite. The heterogeneity of the mineral (mixed with quartz, pyrophyllite, and kaolinite) cannot be used to justify that Cu and Zn are attributable to impurities. The presence of these two elements assumed to substitute for Mn^{2+} in the formula of Ottré davreuxite could be explained by their geochemical behavior during the formation of the quartz veins through the Upper Salmian metapelites of the Stavelot Massif, characterized by a particular chemical composition (Kramm, 1973, 1976, and Fransolet et al., 1977).

Davreuxite requires further petrographic investigation. The relatively frequent association davreuxite–andalusite in the quartz veins must have genetic implications but the formation conditions of this association have not been accurately described. We finally mention that the fibrolite-like aspect of davreuxite in thin section and, therefore, the possible confusion with sillimanite could constitute one of the reasons why the mineral is apparently rare. It is evident that X-ray investigations and/or microprobe analyses must be more systematically conducted on sillimanite-like phases, especially in Mn-rich geological environments, in order to detect davreuxite.

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