Genesis of tourmalinites from Belgium: petrographical and chemical evidence

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ABSTRACT. The petrographical and chemical character-istics of the tourmalinite pebbles from the Lower Devonian conglomerates of Belgium are presented to-gether with selected trace element data. The tourmalinites are shown to represent metasomatically altered rocks of variable origin and composition. A model of subvolcanic hydrothermal alteration is proposed.

THE occurrence of tourmalinites in Belgium is confined to the conglomerates of the Lower Devonian, in which they appear as black pebbles of variable size, shape, and roundness. Their relative abundance varies considerably from one locality to another. The pebbles cannot be traced to the exposed underlying basement and thus appear to be exotic.

The earliest description of a tourmalinite pebble from Belgium dates back over a hundred years (de la Vallée Poussin and Renard, 1877). Since then many authors have referred to these rocks. Their descriptions are nevertheless incomplete as most observations are based on too few samples. In spite of this, many hypotheses about the genesis and the original composition of the tourmalinites have been proposed.

According to Lohest (1909) and Anten (1922) the tourmalinites result from the metamorphism of tourmaline placer deposits. This hypothesis is sup-ported by the banding of several tourmalinite pebbles. Most workers, however, believe that the presence of tourmaline must be related to granitic rocks. Kaisin (1930) and Macar (1948) consider the tourmalinites as pneumatolytic differentiates of a hidden granite intrusion. Corin (1930) and Trève (1976) agree with a granitic origin of the boron but believe that the tourmalinites represent meta-somatically transformed sedimentary rocks and tectonic breccias.

Meisl and Ehrenberg (1968), who studied the tourmalinite pebbles in the Gedinnian conglomer-ates of the Western Taunus (Germany), noted a great similarity with the Belgian tourmalinites and concluded that they constitute transformed sediments. These authors could not choose between a contact pneumatolytic and a regional meta-morphic origin for these rocks.

It is the purpose of this paper to describe the most representative tourmalinite varieties found in Belgium and to discuss some chemical properties of their tourmalines. This sheds some light on the primary composition of the rocks and on the origin of the mechanism of tourmalinization they under-went.

Distribution of the samples

All known exposures of tourmalinites in Belgium are indicated on fig. 1. They belong either to the

Cambrosilurian massifs

Lower Devonian

Younger rocks
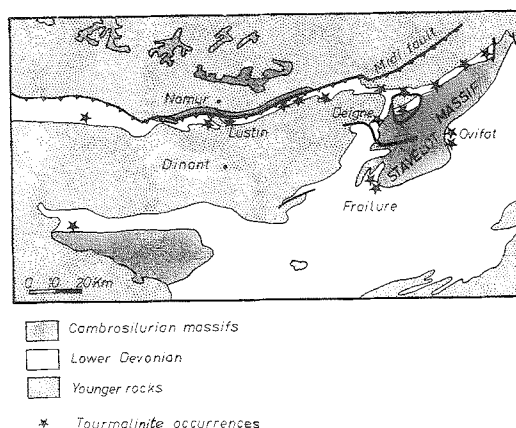
✳   Tourmalinite occurrences

FIG. 1. Geological map of Southern Belgium showing the tourmalinite occurrence.

marine formations of the Gedinnian (Quarreux and Ombret conglomerates) or to the Upper Emsian (Burnot conglomerates). For the present study a few hundred tourmalinite pebbles sampled at four localities were analysed. The rocks from Lustin and Deigne are of Upper Emsian age whereas those from Fraiture and Ovifat form part of the Quarreux conglomerates.

Petrographical description of the main tourmalinite varieties

Tourmaline and quartz are the essential components of tourmalinites. The proportions of both minerals can vary appreciably. If the tourmaline content is very low the rock resembles a tourmaline-bearing quartzite. Even in that case the term tourmalinite will be used for convenience.

Tourmalinites with relicts of igneous textures

Equigranular rocks. Only one pebble belongs to this group. It is a medium- to coarse-grained rock consisting mainly of large equigranular anhedral quartz grains and tourmaline prisms surrounded by numerous slender radiating aggregates of tourmaline. The accessory minerals are apatite, zircon, sphene, and rutile.

The two generations of tourmaline in the rock recall the luxullianite of St. Austell, Cornwall (Wells, 1946; Lister, 1978). A first generation of tourmaline is usually massive prismatic and much embayed by corrosion. Such grains are large and exhibit strong colour zoning. The distribution of the colour zones is very irregular. The core is generally nearly colourless (ε) to greenish brown or yellowish brown (ω). Along the margins the brownish colours are replaced by greenish or indigo hues. These large tourmaline grains are considered to be primary constituents of the tourmalinized rock. They often show overgrowths of secondary tourmaline with radiating habit and small size.

It seems, however, that the bulk of the tourmaline occurring in this rock developed as radiating or spherulitic aggregates of secondary origin. From their shape it can be concluded that some aggregates represent pseudomorphs after feldspar. Except for one crystal the replacement of the feldspar was always complete. Other tourmaline aggregates are feather-like and relatively coarse-grained. Relicts of the cleavage planes of the original mineral are still observable but the mineral itself has completely disappeared. The shape of the latter aggregates as well as the presence of a perfect cleavage suggest that they replaced biotite or another micaceous mineral. This is also supported by the observations of Lister (1978) who noticed the feather-like development of tourmaline replacing biotite in the St. Austell granite. Whatever the composition of the original minerals the colour of the secondary tourmaline is different from that of the primary tourmaline crystals. Indeed, the colour along (ω) is always bottle-green or bluish green in the radiate, stellate, or fibrous aggregates.

The textural properties described above point to a primary granitic composition of the rock. This is also suggested by comparing the petrographical properties of the sample with the characteristics of the St. Austell luxullianites (Wells, 1946).

Porphyritic rocks. This infrequent tourmalinite variety is characterized by quartz phenocrysts, associated with numerous large tourmaline pseudomorphs. The matrix is microgranular and consists of quartz, tourmaline, and subordinate amounts of sericite.

The quartz phenocrysts are corroded or embayed crystals of variable size. Although their average grain size is about 2 mm some porphyritic specimens carry quartz phenocrysts which are more than 5 mm across. Most pseudomorphs formed at the expense of euhedral to subhedral elongated crystals. Some of them are larger than 1 mm. The shape of the tourmaline pseudomorphs suggests that they developed after feldspar and/or after a mafic constituent (biotite?).

Although three types of tourmaline pseudomorph are recognized transitional types are quite common. Most pseudomorphs consist of very fine-grained aggregates of randomly oriented pale green tourmaline needles. Others are composed of a few relatively large idiomorphic tourmaline grains with prominent colour zoning and some interstitial quartz. A third type of pseudomorph is composed of radial aggregates of tourmaline of variable size. The occurrence of abundant rutile inclusions in some of these pseudomorphs may result from the titaniferous nature of the primary mineral.

The matrix of the porphyritic tourmalinites is mainly composed of anhedral quartz grains. Their average grain size is about 0.05 mm. The other components of the groundmass are fibrous, scaly or radiate in habit, and include tourmaline and some sericite. Idiomorphic rutile, zircon, and opaque minerals are the accessories.

The porphyritic tourmalinites may correspond either with tourmalinized microgranites or with acid volcanic rocks.

Hydrothermal breccias

The brecciated tourmalinites form a moderately abundant group. The size of the rock fragments is highly variable; some of them are more than 2 cm across. The fragments are unsorted and their shape
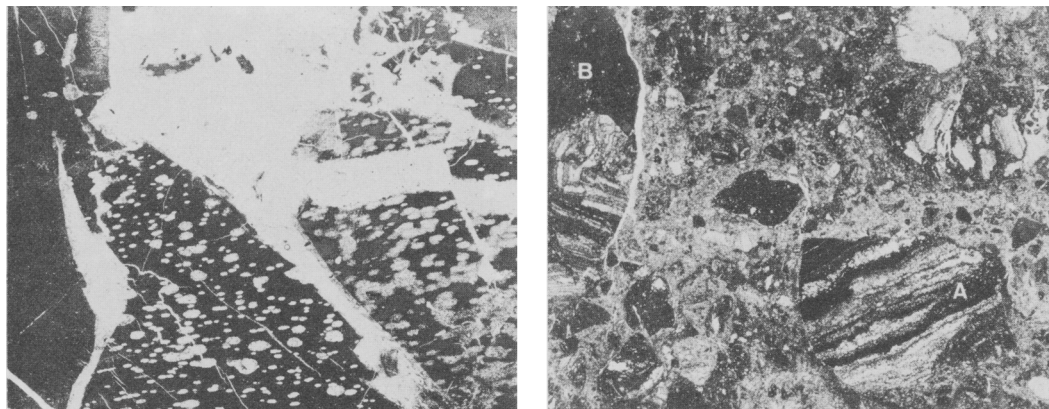
FIGS. 2 and 3. FIG. 2 (*left*). Hydrothermal breccia L3 with fragments showing spherical or ovoid bodies; quartz is white, tourmaline is black. FIG. 3. (*right*). Altered volcanic breccia L12 with banded (A) and brecciated (B) rock fragments (one nicol, × 3).

is angular to subangular. Several kinds of breccia are distinguished according to the nature of the rock fragments. In some breccias all fragments initially consisted of the same parent material. More frequently materials of different origin and composition are mixed.

The original mineralogical composition of the different rock fragments is always obscured by the tourmalinization. Nevertheless, thus it appears that most of them are of volcanic origin. The volcanic nature is suggested by the following features: the presence of pseudomorphs after phenocrysts; the occurrence of spherical or ovoid bodies resembling cavity fillings or vesicles (fig. 2); the shard-like shape of some rock fragments; the subparallel arrangement of some tourmaline crystals suggesting a flow structure; some samples contain fragments derived from volcanic breccias (fig. 3). An intermediate to basic composition for the volcanic fragments is likely as they never contain quartz phenocrysts. The tourmalinization of the volcanic material always resulted in the complete replacement of the primary minerals by tourmaline and subordinate quartz. Whereas the phenocrysts are replaced by relatively large green-brown tourmaline, the groundmass of the volcanic fragments always consists of very fine-grained tourmalines of darker shades.

Some breccias also contain quartzitic inclusions (fig. 4) or banded rock fragments with alternating tourmaline-rich and very fine-grained quartz layers. Some of these fragments certainly correspond with tourmalinized sediments but a pyro-clastic or volcano–detrital origin is more likely for others (cf. banded tourmalinites).

The matrix of most breccias is a mixture of equigranular to elongated anhedral quartz grains,

tourmaline needles, and small angular rock fragments. It is usually enriched in quartz, whereas the fragments consist preferentially of tourmaline. The quartz matrix of a few breccias encloses slender prismatic apatite crystals. In one tourmalinite pebble several apatite crystals are more than 1 cm long. Rutile and iron oxides are the main accessories.

It is undeniable that the brecciated character of most rocks of this group is also partly due to a chemical brecciation (Sawkins, 1969) (fig. 5). This brecciation resulted from intense hydrothermal processes which affected the rocks and induced the partial or complete replacement of the primary minerals by tourmaline and quartz. All rocks of this group are therefore considered as hydro-thermal breccias.

Banded tourmalinites

Such tourmalinites form the most widespread group. Depending on the development of the banding, at least three subgroups may be recognized.

A first subgroup includes tourmalinites which look megascopically as greyish black to black aphanitic igneous rocks. The banding is due to extremely thin layers of splinter-like quartz crystals. The remaining part of the rock is composed of a dark indistinctly crystalline matrix of green tourmaline. Sporadically these rocks enclose large rectangular quartz-tourmaline pseudomorphs. They are up to 3 mm long and their shape suggests that they replace phenocrysts of the primary rock. Some of these pseudomorphs include numerous idiomorphic rutile grains associated with opaque minerals. Some tourmalinites also contain abundant ovoid or spherical bodies with an average
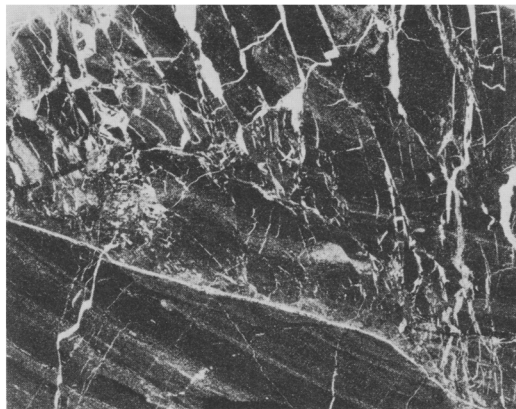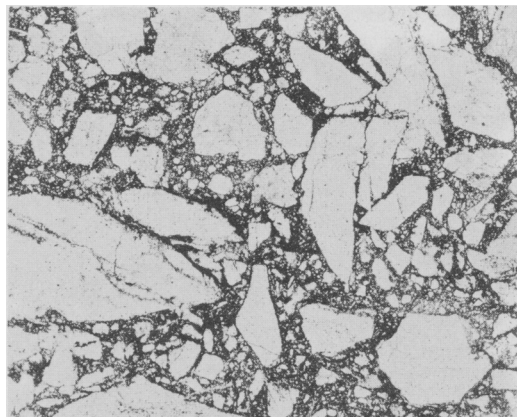
FIGS. 4 and 5. FIG. 4 (*left*). Hydrothermal breccia L28 consisting of quartzitic fragments (white) set in a fine-grained tourmaline matrix (black) (×2.5). FIG. 5 (*right*). Hydrothermal breccia L22 mainly composed of finely banded rock fragments (one nicol, ×0).

diameter of 0.6 to 0.8 mm. They are filled up by a mixture of anhedral quartz, prismatic tourmaline, and idiomorphic opaque grains.

The second subgroup is characterized by a more prominent banding. Most layers are lenticular and consist of anhedral quartz with some tourmaline, sericite, and chlorite. The inner part of these lenses usually resembles a quartz mosaic. Between the quartzitic layers the rock consists of a nearly monomineralic aggregate of microcrystalline tourmaline. Several tourmalinites of this subgroup include annular siliceous bodies. The rings are 0.05 to 0.12 mm across and may enclose a few small tourmaline grains. Some of these annular bodies are squeezed completely flat. In extreme cases the whole lens is built of such distorted rings.

In a third subgroup the fine-grained quartz-rich layers are more than 5 mm thick and enclose scattered, irregular patches of·xenomorphic tourmaline and interstitial sericite. Between them are observed tourmaline-rich layers of variable thickness, composed of xenomorphic tourmaline in which small rounded poikilitic quartz crystals and interstitial sericite are embedded. Although the grain size of the quartz is comparable with that of the tourmaline grains, lateral and vertical variations may occur within one single bed or between different beds.

In none of the banded tourmalinites is a relict of a possible primary schistosity observed. As related above it is suspected that at least some banded tourmalinites are of pyroclastic origin or correspond to volcanic sediments. This applies particularly to the rocks of the first subgroup. The occurrence of sedimentary structures (cross-bedding and load-casting) in some tourmalinites

of the third subgroup, on the other hand, leaves the origin of these rocks beyond doubt.

Tourmalinites without relict textures

All tourmalinites which are devoid of relict textures are for convenience grouped under this heading. Their major constituents are tourmaline, quartz, and sericite. These minerals appear in varying proportions but sericite is a more important component here than in the varieties described above. Nevertheless there is never more than 5 % in volume of this mineral. The fabric and the average size of the tourmalinites of this group may not only vary drastically from one sample to another but also within a single hand specimen.

Some coarse-grained samples are almost pure tourmaline-bearing rocks in which the texture is hypidiomorphic-granular to allotriomorphic-granular. The majority of the large tourmaline crystals shows deformation and granulation. Indeed, many tourmalines exhibit undulatory extinction, complicated internal gliding structures, and recrystallization phenomena. The few irregular patches between adjacent tourmaline prisms are filled with quartz, sericite, and minute tourmaline needles. Rutile and opaque minerals are the only accessory minerals found in this kind of tourmalinite. The veinlets which cut the rock are mainly composed of anhedral quartz which contains acicular to prismatic tourmaline needles.

With increasing quartz content several textural varieties of tourmalinite are recognized. In some samples the tourmaline crystals are embedded in a matrix of sericite and poikilitic quartz. In others, when tourmaline and quartz occur in approximately equal proportions, large anhedral quartz

grains constitute the groundmass. Some quartz grains are more than 1 mm across and enclose several tourmaline crystals. The accessory minerals are few and consist of rutile and iron oxides.

Tourmalinites containing less than 25% tourmaline usually consist of interlocking anhedral quartz grains of variable size. Irregular or hypidiomorphic tourmaline crystals are located at the contact of the quartz grains as well as inside them. In some of these quartzitic rocks the tourmaline grains are arranged in a subparallel manner. Muscovite or sericite aggregates surround some quartz crystals and it seems that tourmaline is often partially converted into sericite. Zircon, rutile, and sphene are the main accessories of these quartzitic tourmaline-bearing rocks.

Chemical data

Methods

After crushing, all samples were treated with aqua regia in order to eliminate iron oxide before analysis. Microscopic examination of four tourmaline separates ensured that all impurities had been removed.

The major element analyses were carried out using the standard procedure of Van Hende (1976). B_2O_3 was determined by the colorimetric method of Dietz (1975). FeO determinations resulted from ^{57}Fe Mössbauer spectrometry, Cu, Zn, Pb, Ni, Sn, and Co were analysed by atomic absorption spectrometry and W was determined by means of the colorimetric method of Storms et al. (1980). Rb, Sr, Zr, and Y were determined by X-ray fluorescence.

Results and interpretation

Table I gives the major element chemistry of the samples. The data indicate that the tourmalines belong to the isomorphous schorl-dravite series. The uvite component is always negligible and all analysed tourmalines have FeO > Fe_2O_3.

Vladykin et al. (1975) concluded from a compilation of more than 300 tourmaline analyses that

TABLE I. *Chemical analyses of tourmalines and tourmalinites from Belgium*

	1* (%)	2* (%)	3* (%)	4* (%)	5 (%)	6 (%)	7 (%)
SiO$_2$	35.12	34.17	34.85	35.08	71.62	61.05	48.04
Al$_2$O$_3$	31.22	31.47	34.22	35.18	14.66	20.35	27.85
B$_2$O$_3$	10.24	10.56	11.09	10.70	4.08	5.79	8.19
Fe$_2$O$_3$	3.07	4.84	0.51	0.93	0.96	0.57	0.51
FeO	8.27	9.27	7.20	11.10	3.68	5.91	6.08
MgO	4.83	4.20	6.08	1.99	1.25	1.37	3.71
CaO	0.81	0.47	0.54	0.37	0.12	0.18	0.23
Na$_2$O	1.78	2.02	1.74	1.22	0.58	0.70	1.34
K$_2$O	0.02	0.01	0.02	0.03	0.01	0.08	0.25
MnO	0.06	0.04	0.06	0.04	0.03	0.02	0.04
TiO$_2$	1.08	0.26	0.52	0.45	0.48	0.98	0.63
H$_2$O	3.52	3.41	3.27	3.77	2.39	3.20	3.57
Total	100.02	100.72	100.10	100.88	99.86	100.20	100.44

Number of ions

	1*		2*		3*		4*		5	6	7
Si	5.868		5.712		5.719		5.777				
B	2.952		3.046		3.139		3.042		2.717	2.796	2.870
Al	6.000		6.000		6.000		6.000		6.000	6.000	6.000
Al	0.150		0.202		0.657		0.830		0.666	0.713	0.665
Fe$_{3+}$	0.386		0.609		0.063		0.115		0.278	0.119	0.078
Mg	1.203		1.047		1.487		0.489		0.719	0.572	1.122
Ti	0.136		0.033		0.064		0.055		0.139	0.207	0.096
Fe$_{2+}$	1.156	3.764	1.296	3.934	0.988	3.920	1.529	3.483	1.187	1.323	1.032
Mn	0.008		0.006		0.008		0.006		0.009	0.005	0.007
Na	0.576		0.655		0.554		0.390		0.434	0.380	0.527
Ca	0.145		0.084		0.095		0.065		0.049	0.054	0.050
K	0.004		0.002		0.004		0.004		0.005	0.029	0.065
OH	3.923		3.803		3.579		4.140		3.074	2.986	2.416

Analyst: J. Van Hende (RUG).

* Pure tourmaline separates; number of ions based on 31 (O,OH).

† Rock analyses; as the samples consist of quartz-tourmaline mixtures, the number of ions was calculated assuming a sum of 9 ions per fomula unit for the elements Al, Fe, Mg, Ti, and Mn.

Analysis 1(L40), 2(L4), 3(FO), and 7(F6) represent tourmalinites without relict textures; 4(LI), 5(L12), and 6(LO) represent respectively a porphyritic type, a tourmalinized volcanic breccia, and a banded tourmalinite of the second subgroup.

four main genetic groups of tourmaline occur in nature. Each of these groups corresponds to a distinct field in the triangle representing the number of R ions (Mg–Al, Li–Fe^{2+}, Fe^{3+}, Mn, Ti, recalculated to 100%) in the tourmaline, assuming a general structural formula of NaR_3Al_6 Si_6O_{18} $(BO_3)_3(OH,F)_4$ as given by Deer et al. (1962). According to Vladykin et al. (1975) the dravites (group IV) are usually found in metamorphic and carbonate rocks. In this respect it is important to mention that Agrell (1941) and Deer et al. (1962) noticed that the Mg-rich character of the dravite is controlled by the chemical composition of the host rock. Dravite was also recorded by Abraham et al. (1972) from tourmalinized metasediments of the Arzberg series in West Germany. Magnesian schorls (group III) are characteristic of muscovite pegmatites associated with metamorphic belts. Granites and their associated pegmatites contain iron-rich schorls (group II). Power (1968) and Neiva (1974) showed that the chemistry of these tourmalines appears to be related to the stage of magmatic fractionation of the granitoid rocks in which they occur. Certain quartz–tourmaline rocks discussed by Power (1968) are shown to be the product of extreme fractionation. Finally, group I of Vladykin et al. (1975) includes the elbaites crystallizing in Na–Li pegmatites.

The tourmaline groups distinguished by Vladykin et al. (1975) together with the plots of all samples listed in Table I are shown in fig. 6 which leads to the following conclusions. The four tourmaline samples separated from rocks without relict textures plot into the field of the muscovite pegmatites (group III). They show no similarity with the quartz–tourmaline rocks discussed by

Power (1968) but their composition is intermediate between schorl and dravite. It is obvious that the chemical data are strongly influenced by the zoning of the tourmaline crystals. Semiquantitative microprobe data along traverses across these crystals revealed increasing Mg and Na contents towards the rims at the expense of Fe, Al, and Ti. From the petrographic characteristics and from the chemistry of the tourmaline samples belonging to group II we conclude that the tourmalinites analysed derive from late-stage vein deposits.

The tourmalines which characterize the volcanic breccia and the porphyritic igneous rock have a schorlitic composition as both plot in the field II of fig. 2. This agrees well with literature data on tourmalines from rocks of identical origin.

Why the tourmaline in the banded rock of the second subgroup also plots in the same field (field II) is less obvious. If the tourmaline results from the metamorphism or the metasomatism of a pelitic rock, a dravitic composition would be expected. As the mineral is a schorl with a chemical composition very close to that of the tourmalines found in the porphyritic rock and in the volcanic breccia, this can be an argument in support of the pyroclastic nature of the primary rock. This deduction is very speculative, however, as we are unable to evaluate the nature of the metasomatizing solutions.

Data for selected trace elements of total rock samples are presented in Table II. No clear relationship is found between the tourmalinite variety and its trace element content. All samples analysed contain normal concentrations of the metallogenetic elements Cu, Zn, Pb, Ni, and Co. Sn could not be detected and the W content is usually lower than 10 ppm. Only one sample contains a much higher tungsten content (F 102 ppm). It corresponds with a tourmalinite pebble which consists of one large quartz crystal with abundant inclusions of needle-like tourmaline.

Origin of the tourmalinites

The petrographic study of the tourmalinite pebbles from the Lower Devonian of Belgium supports a metasomatic origin for these rocks. Although the metasomatic alteration affected rocks of variable origin and composition, it was established that at least a part of them are of magmatic origin. Among them volcanic, subvolcanic, and pyroclastic rocks are predominant.

Some of these rocks were subjected to a pervasive brecciation which may be related to the passage of hydrothermal solutions, which would substantially obscure the texture and the primary mineralogical composition of the rocks. Brecciated
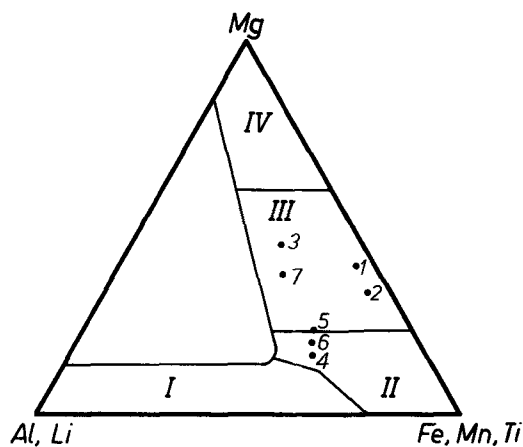

FIG. 6. Triangular diagram after Vladykin et al. (1975). Numbers refer to chemical analyses presented in Table I.

TABLE II. *Trace element content (ppm) in tourmalinites from Belgium*

Rock variety	Sample no.	Zn	Pb	Cu	Ni	Co	Rb	Sr	Zr	Y
Porphyritic	D1	8	7	14	7	7	6	40	149	23
	L1	12	14	14	5	4				
Brecciated	D2	8	15	10	26	18				
	L2	6	7	3	6	5				
	L9	22	5	15	7	4				
	L12	14	9	23	17	11	n.d.	83	95	28
	L14	10	4	4	7	8	n.d.	28	37	20
	L15	14	14	15	6	6				
Banded	L0	10	52	10	13	7	2	105	183	58
	L3	16	14	26	11	13	7	77	120	36
	L5	14	9	4	12	13	56	30	247	17
	L6	7	14	51	16	3				
	L11	24	7	51	13	9	2	173	178	27
	L13	10	22	23	12	12				
	L16	19	18	7	15	9	3	67	185	40
	L17	12	12	12	17	16				
	NR20	15	45	3	21	20				
Undifferentiated	L4	9	9	5	15	11				
	L40	13	9	6	17	13				
	NR1	5	9	n.d.	4	3				
	NR6	13	12	2	7	7	15	187	104	8
	O1	8	7	2	6	5				

Analysts: L. Buvé (Zn–Pb–Cu–Ni–Co).
  M. Delvigne and F. Durez (Rb–Sr–Zr–Y).
n.d.: not detected.

tourmalinites originating in tectonic processes were not found.

The recurrent presence of altered igneous rocks among the tourmalinite samples is considered as a highly relevant property suggesting a close relationship between the magmatism and the subsequent alteration of the rocks.

A model of subvolcanic alteration which explains the occurrence of porphyry tin deposits in Bolivia has been proposed by Sillitoe et al. (1975). This model seems also applicable to the tourmalinites discussed here. According to that model, intrusions of quartz porphyries, brecciation, alteration, and porphyry-type and vein mineralization comprise a single sequence of cogenetic events (Sillitoe et al., 1975). In many respects the pervasive quartz–tourmaline alteration of the volcanic deposits located in the core of the Chorolque centre (Bolivia) is comparable with that observed in the tourmalinites under consideration. Some differences nevertheless exist but in our opinion, taking into account the mechanism involved, they are not essential. The main objection to the application of this ortho-

magmatic model is perhaps the absence of mineralization in the tourmalinite samples as shown by the results of the trace element analyses. The occurrence of lode systems (Sn–W–Ag) is highly characteristic of the Eastern Cordillera of the Central Andes indeed, but they are certainly not omnipresent in the whole region of the Andes affected by the hydrothermal alteration and brecciation.

The textural characteristics of the brecciated tourmalinites from Belgium also recall some aspects of the tourmaline-cemented intrusive breccias of SW England, described by Goode and Taylor (1980). Definite conclusions are, however, unjustified since we have no indication of the geological environment from which the pebbles derived.

out by Mrs Buvé in the laboratories of Professor Viaene (KUL). Dr Delhal (KMMA) supplied the X-ray fluorescence data, Dr De Grave (RUG) the Mössbauer spectrometry analyses, and Professor Laduron (UCL) supervised the microprobe runs. Our special thanks to Professor Viaene (KUL) for critical reading of the manuscript and to Professor King (KUL) for improving our English text.

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