

CHEMICAL COMPOSITION OF TOURMALINE IN QUARTZ VEINS FROM NEA RODA AND THASOS AREAS IN MACEDONIA, NORTHERN GREECE

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ABSTRACT

Crystals of tourmaline from quartz veins cross-cutting metamorphic rocks from Nea Roda (Chalkidiki Peninsula) and Thimonia (Thasos Island) in Macedonia (northern Greece) display optical zoning (bluish green to brownish green) and core-to-rim chemical variations. In both cases, the tourmaline belongs to the schorl–dravite solid-solution series, with that from Thasos being more dravitic, and presenting a limited extent of coupled substitution toward an alkali-free or proton-deficient member. The chemical composition of the tourmaline suggests that the host quartz veins are not related to nearby granites. Field observations and textural features indicate deposition prior to the latest episode of metamorphism and deformation of the surrounding metasedimentary rocks. The tourmaline retained its primary composition and zonal texture in spite of the metamorphic overprint.

Keywords: tourmaline, schorl–dravite, quartz veins, zoning, Macedonia, Greece.

SOMMAIRE

Des cristaux de tourmaline provenant de veines de quartz recoupant un socle métamorphique des régions de Nea Roda (péninsule de Chalkidiki) et de Thimonia (île de Thassos), en Macédoine, dans le nord de la Grèce, font preuve d'une zonation optique (vert bleuâtre à vert brunâtre) et chimique, du coeur vers la bordure. La tourmaline des deux endroits fait partie de la série de schorl à dravite, l'exemple de Thasos étant davantage dravitique; par contre, elle montre une légère déviation vers le pôle sans alcalin ou déficitaire en protons. D'après sa composition, les veines de quartz qui la renferme ne seraient pas liées aux granites du milieu. Les relations de terrain et le développement textural indiquent une déposition antérieure au dernier stade de métamorphisme et de déformation des roches métasédimentaires encaissantes. La tourmaline a conservé sa composition primaire et sa zonation malgré cet épisode de métamorphisme.

(Traduit par la Rédaction)

Mots-clés: tourmaline, schorl–dravite, veines de quartz, zonation, Macédoine, Grèce.

INTRODUCTION

Because of its wide range of stability, tourmaline is a common accessory mineral in a variety of geological environments. Its composition can provide useful information about the environment and conditions of formation (*e.g.*, Manning 1982, Henry & Guidotti 1985, Jolliff *et al.* 1986, Kassoli-Fournaraki 1990) and the bulk chemistry of the system in which it forms (*e.g.*, Peltola *et al.* 1968, Ethier & Campbell 1977, Ayuso & Brown 1984, Gallagher 1988). Also, because it is one of the chemically most resistant minerals, it has the potential for preserving primary chemical features through multiple hydrothermal and metamorphic events.

In this paper, we present chemical and textural data concerning tourmaline in quartz veins cropping out near Nea Roda on the isthmus of Athos peninsula (Chalkidiki), and near Thimonia on the southern part of Thasos island (Fig. 1), and provide a petrogenetic interpretation of this type of occurrence.

LOCATION AND GEOLOGICAL SETTING

The Nea Roda area belongs to the Vertiskos Group of the Serbomacedonian Massif. This group is characterized by a complicated history of tectonometamorphic evolution (*e.g.*, Kockel *et al.* 1977, Kassoli-Fournaraki *et al.* 1985, Sakellariou 1989, Sidiropoulos 1991).

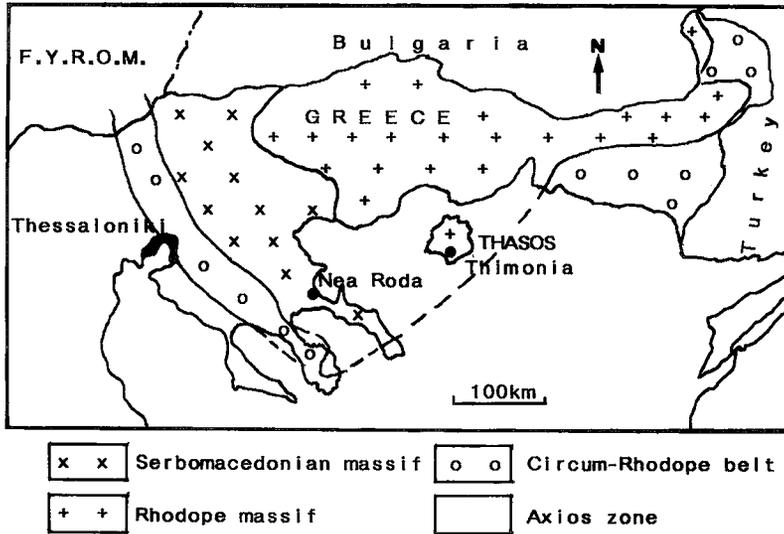


FIG. 1. Simplified geotectonic map of a part of northern Greece, showing locations of the areas studied. F.Y.R.O.M.: Former Yugoslavia Republic of Macedonia.

Briefly, the region is composed largely of migmatitic gneiss, mica schist, amphibolite, a fault-bounded serpentinitized ultramafic body and Neogene sediments. Tertiary granites, granodiorites and associated pegmatite veins have intruded in the area (De Wet *et al.* 1989). Tourmaline-bearing quartz veins, ranging in thickness from a few cm to more than one meter, cross-cut the migmatitic gneiss.

The island of Thasos belongs to the Rhodope Massif and consists of a series of interbedded metasedimentary rocks including marble (calcitic or dolomitic), schist and gneiss. The last deformation and regional metamorphism of Thasos are considered to be related to the Mesozoic – Early Tertiary Alpine orogeny. A sedimentary character for the precursor rocks was determined geochemically, and a low- to medium-grade (Vavelidis *et al.* 1988) or even high-grade (sillimanite zone, Dimitriadis 1989) metamorphism has been demonstrated for the Thasos metamorphic suite. Tourmaline–quartz veins of diverse dimensions occur within sericite–chlorite schists of this sequence. The tourmaline-bearing bodies and the enclosing schists have been folded and locally fractured.

PETROGRAPHY

Nea Roda

Needle-like (up to 5 cm long) or prismatic isolated euhedral to subhedral black crystals of tourmaline occur in massive quartz in the Nea Roda veins. The

modal concentration of tourmaline does not exceed 10% by volume, and its distribution in the veins is quite uniform. Fracturing of the needle-like crystals is commonly attributed to deformation accompanying metamorphism; quartz heals the fractures (Fig. 2a). Inclusions in tourmaline or quartz–tourmaline intergrowths are present but scarce. Traces of the feldspars and white mica occur sporadically in the quartz veins.

Most of the grains of tourmaline display two optically distinct growth-zones (Figs. 2a, b), which are developed concentrically about the *c* axis and nearly parallel to the crystal faces. A light blue-green core gives way to a darker brownish green rim. Optically homogeneous (*i.e.*, unzoned) crystals are rare.

Irregular zoning occurs as patchy domains of blue-green in brownish green regions. Some well-defined cores have outer zones showing an irregular distribution of coloring (Fig. 2b).

Thasos

Sprays of black prismatic crystals or clots of composite aggregates of tourmaline occur in the quartz veins on Thasos. Modal tourmaline exceeds 50% by volume; thus, the term tourmalinite veins can be used in the sense of Nicholson (1980) and Fortey & Cooper (1986). Euhedral to subhedral and skeletal (incompletely formed) crystals and intergrowths with the associated silicates also are present. In most cases, the development of these intergrowths did not significantly affect either the euhedral character (outer

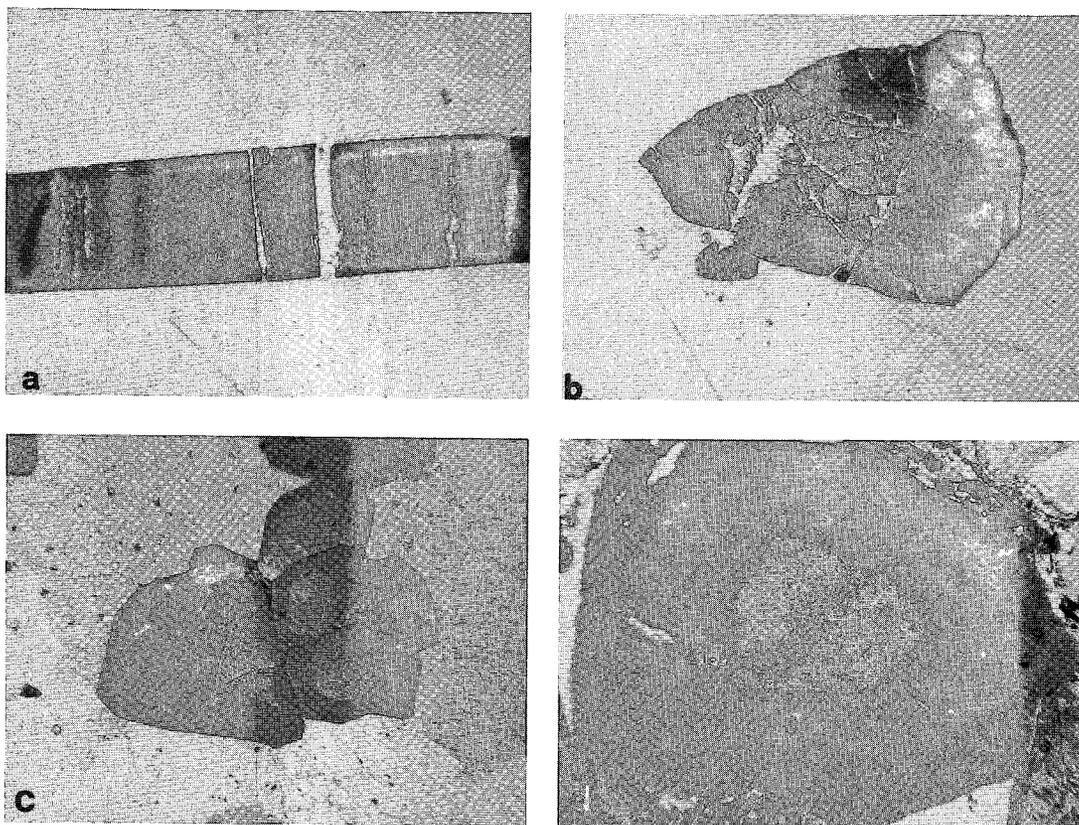


FIG. 2. Photomicrographs (plane-polarized light) showing the textural features of the tourmaline and associated minerals. a) A needle-like zoned crystal of tourmaline, with fractures infilled by quartz (white). Width of the field of view is 2 mm. b) Zoned crystal of tourmaline with a light core and outer zones showing intermixed colors, in a matrix of quartz (white). Width of the field of view is 2 mm. c) Polygonal aggregate of zoned crystals of tourmaline in a matrix of quartz (partly crossed nicols). Width of the field of view is 2 mm. d) Zoned crystal of tourmaline, with several distinct color zones. Width of the field of view is 3 mm.

crystal faces) or the concentric growth-zoning of the individual crystals (Fig. 2c).

Tourmaline grains from Thasos may be unzoned, irregularly zoned or have two sharply defined zones. Color zonation is characterized by a light bluish green core and a deep green rim. Some crystals have several distinct zones (Fig. 2d).

Associated minerals may include quartz and minor sericite, chlorite, sericitized plagioclase and titanite. Martite with exsolution lamellae of titanite after ilmenite is the major opaque phase in the samples studied.

Tourmaline also occurs sporadically as disseminated grains within the sericite–chlorite schist wall-rocks.

Some microtextural relations indicate that tourmaline formed prior to the latest deformation and regional metamorphic events that affected the sur-

rounding lithologies. Evidence in support comes from the following textural features: a) at hinges of microfolds, tourmaline is fractured and bent and has strained extinction; b) fractured grains of tourmaline have their fractures healed by later-formed quartz and chlorite; c) the foliation, defined by chlorite and sericite and wrapping around tourmaline prisms or aggregates, can be considered as metamorphic, although contemporaneous tourmaline or tourmaline postdating metamorphism may also display foliation.

TOURMALINE CHEMISTRY

Analytical procedures

Analyses were carried out using a JEOL JSM-840 scanning electron microscope equipped with a LINK AN 10000 EDS microanalyzer. Operating conditions

were: accelerating voltage 15 kV, beam current 3 nA, beam diameter 1 μm , and counting time 100 seconds. No evidence of beam damage was observed. Corrections were made using the ZAF-4/FLS software provided by LINK. Natural minerals or synthetic equivalents (albite, orthoclase, diopside, olivine, corundum, periclase, apatite, barite and pyrite) and pure metals were used as microprobe standards.

The analytical precision was found to be variable for different elements (average values in parentheses): Si (0.11), Al (0.12), Ti (0.06), Fe (0.17), Mg (0.13), Mn (0.07), Ca (0.05), K (0.04), Ni (0.13), Cr (0.07), V (0.06), Zn (0.20), Pb (0.09), Sr (0.14), Cu (0.14), Na (0.40), and Ga (0.27); some of them were at or below the limit of detection.

Optically zoned and unzoned crystals were analyzed along rim – core – rim traverses. Representative compositions of rim, core and intermediate zones (where present) are shown in cation plots to express chemical variations. Selected compositions of zoned tourmaline grains (one grain from each area) and cal-

culated cation formulae based on 24.5 oxygen atoms are shown in Table 1. All iron is considered to be present as Fe^{2+} , and B was assumed to obey stoichiometry requirements.

Element concentrations

The tourmaline crystals analyzed display SiO_2 variation within the range of 36.6–39.3 wt% and 37.3–38.9 wt% for Nea Roda and Thasos, respectively. Analogous variation is displayed by Al_2O_3 , with a range of 29.5–32.1 wt% and 30.4–33.7 wt% for Nea Roda and Thasos, respectively. FeO ranges between 7.2 and 9.8 wt% (Nea Roda) and between 3.9 and 5.9 wt% (Thasos), whereas MgO ranges between 6.9 and 7.8 wt% (Nea Roda) and between 8.0 and 9.2 wt% (Thasos).

The electron-microprobe analyses show that the tourmaline of both study areas belongs to the schorl–dravite solid-solution series. The tourmaline from Thasos is more dravitic than that of Nea Roda,

TABLE 1. RESULTS OF SCANNING ELECTRON-MICROPROBE ANALYSES OF REPRESENTATIVE GRAINS OF TOURMALINE FROM NEA RODA AND THASOS AREAS, MACEDONIA

	Nea Roda – Grain NR3						Thasos – Grain Th4					
	rim	core		rim	rim	int	core	int	rim			
SiO_2 wt%	36.36	36.79	37.09	37.26	36.97	37.49	37.42	37.52	38.81	37.72	37.62	
Al_2O_3	29.18	30.13	30.45	30.88	28.75	30.11	30.97	31.62	33.59	31.58	30.36	
TiO_2	1.03	0.54	0.09	0.12	1.07	0.72	0.29	0.27	0.13	0.48	0.54	
FeO	9.12	7.99	7.96	8.11	9.31	8.11	5.53	5.24	3.81	5.69	5.69	
MgO	7.10	7.28	7.71	7.60	6.86	7.32	8.71	8.51	8.48	8.73	9.20	
MnO	0.00	0.06	0.05	0.00	0.03	0.06	0.10	0.11	0.12	0.02	0.00	
CaO	1.09	0.94	0.30	0.54	1.05	1.10	1.18	0.58	0.14	1.05	1.35	
Na_2O	2.41	2.85	2.17	0.80	2.07	0.92	1.92	3.04	0.55	1.09	1.48	
K_2O	0.07	0.00	0.04	0.00	0.03	0.00	0.05	0.00	0.00	0.00	0.00	
NiO	0.00	0.04	0.23	0.00	0.15	0.05	0.00	0.09	0.00	0.14	0.00	
Cr_2O_3	0.09	0.03	0.00	0.07	0.11	0.00	0.09	0.00	0.05	0.00	0.00	
V_2O_5	0.08	0.02	0.02	0.28	0.21	0.09	0.00	0.14	0.04	0.06	0.00	
ZnO	0.05	0.09	0.23	0.00	0.12	0.00	0.00	0.07	0.11	0.14	0.28	
PbO	0.00	0.02	0.00	0.00	0.00	0.19	0.00	0.00	0.05	0.02	0.00	
SrO	0.00	0.03	0.25	0.00	0.09	0.00	0.00	0.00	0.00	0.00	0.14	
CuO	0.00	0.00	0.00	0.00	0.00	0.19	0.27	0.00	0.00	0.00	0.00	
Ga_2O_3	0.32	0.00	0.81	1.03	0.16	0.75	0.53	0.01	1.04	0.75	0.43	
Total	86.90	86.86	87.39	86.73	87.00	86.10	87.08	87.25	86.97	87.19	87.09	
Cations on the basis of 24.5 [O]												
Si_x	6.011	6.041	6.054	6.074	6.101	6.115	6.043	6.030	6.133	6.039	6.071	
Al_x	5.686	5.831	5.859	5.934	5.592	5.788	5.895	5.990	6.000	5.960	5.776	
Al_x^{IV}	–	–	–	–	–	–	–	–	0.256	–	–	
Fe	1.262	1.098	1.087	1.105	1.286	1.107	0.748	0.705	0.504	0.761	0.769	
Mn	0.000	0.009	0.007	0.000	0.004	0.009	0.014	0.015	0.016	0.003	0.000	
Mg	1.749	1.782	1.876	1.848	1.687	1.780	2.026	2.039	1.998	2.083	2.215	
Ti	0.128	0.067	0.012	0.014	0.133	0.089	0.036	0.033	0.016	0.037	0.065	
Ni	0.000	0.006	0.030	0.000	0.020	0.006	0.000	0.011	0.000	0.018	0.000	
Cr	0.012	0.005	0.000	0.009	0.015	0.000	0.012	0.000	0.006	0.000	0.000	
V	0.009	0.003	0.002	0.030	0.022	0.009	0.000	0.015	0.004	0.006	0.000	
Ga	0.034	0.000	0.085	0.108	0.017	0.079	0.055	0.001	0.105	0.070	0.044	
Zn	0.006	0.011	0.028	0.000	0.014	0.000	0.000	0.008	0.013	0.016	0.034	
Cu	0.000	0.000	0.000	0.000	0.000	0.024	0.033	0.000	0.000	0.000	0.016	
Y total	3.200	2.981	3.127	3.115	3.198	3.103	2.924	2.827	2.918	3.014	3.178	
Ca	0.194	0.166	0.052	0.094	0.186	0.192	0.205	0.100	0.024	0.182	0.233	
Na	0.772	0.909	0.687	0.252	0.664	0.291	0.609	0.948	0.170	0.340	0.462	
K	0.014	0.000	0.008	0.000	0.005	0.000	0.010	0.000	0.000	0.000	0.000	
Pb	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.003	0.001	0.000	
Sr	0.000	0.003	0.023	0.000	0.009	0.000	0.000	0.000	0.000	0.000	0.000	
X total	0.980	1.079	0.770	0.346	0.864	0.491	0.824	1.048	0.193	0.523	0.695	

which is intermediate in composition between dravite and schorl, with a slight tendency toward the schorl end-member.

Minor elements detected in the suite of tourmaline studied include K, Mn, Ni, Cr, V, Zn, Ga and Pb. Among these, Ga displays the highest values, reaching 1.03 and 1.33 wt% Ga_2O_3 for Nea Roda and Thasos, respectively.

COMPOSITIONAL VARIATIONS

A plot of $R^+ + R^{2+}$ versus R^{3+} (Fig. 3) indicates that the rim and intermediate zones of the tourmaline closely approach the ideal schorl–dravite composition (Foit & Rosenberg 1977), whereas the core reflects a limited extent of coupled substitution toward alkali-free or proton-deficient tourmaline. The extent of such substitution may depend on temperature (Foit & Rosenberg 1977, Manning 1982), increasing with

decreasing temperature of crystallization. Without knowledge of the H_2O content and other elements not sought (*e.g.*, B, F, Cl), it is not possible to assess accurately the true degree of proton deficiency or alkali-defect substitutions.

The pattern of element distribution within single grains of tourmaline differs from sample to sample, and even among grains of a single sample. Some zoning, however, is present in nearly all grains. Rim–core–rim traverses show that Mg, Fe, Ca and Ti tend to exhibit a slight increase from core to rim. Aluminum displays an apparent enrichment in cores and depletion in rims. Silicon displays considerable core-to-rim differences, even in grains of the same thin section, but with no characteristic trend. Some of the tourmaline grains from Thasos, however, display a slight Si-enrichment of the rim compared to the core. Characteristic discontinuities for nearly all elements (especially Na) occur within core and rim zones or in

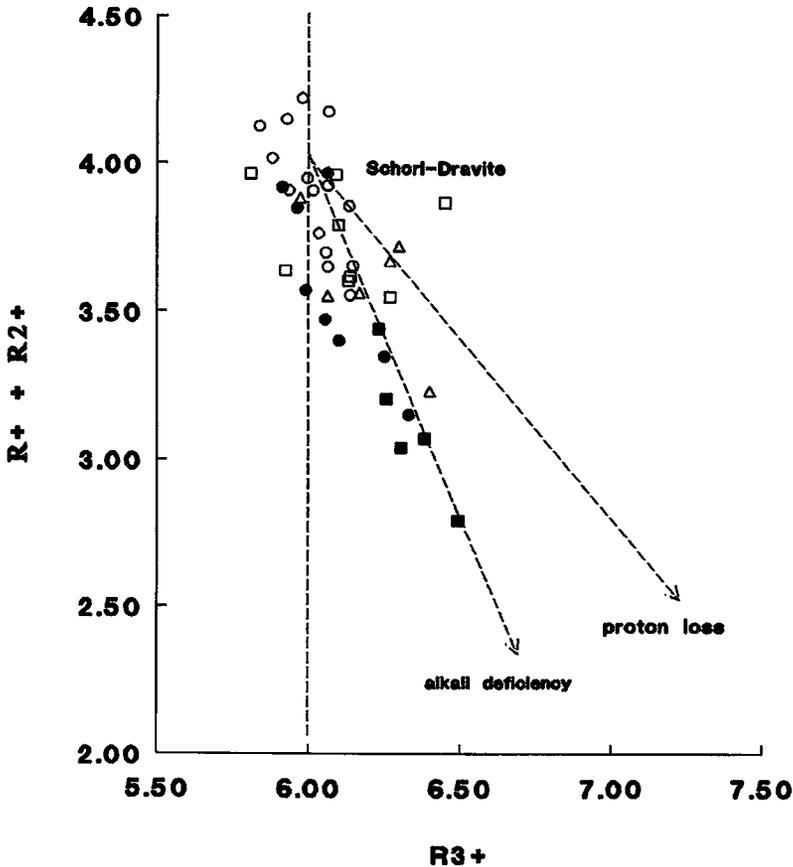


FIG. 3. Alkali-deficiency versus proton-loss substitutions in tourmaline from Nea Roda (filled circle: core, open circle: rim) and Thasos (filled square: core, open square: rim, triangle: interior zone).

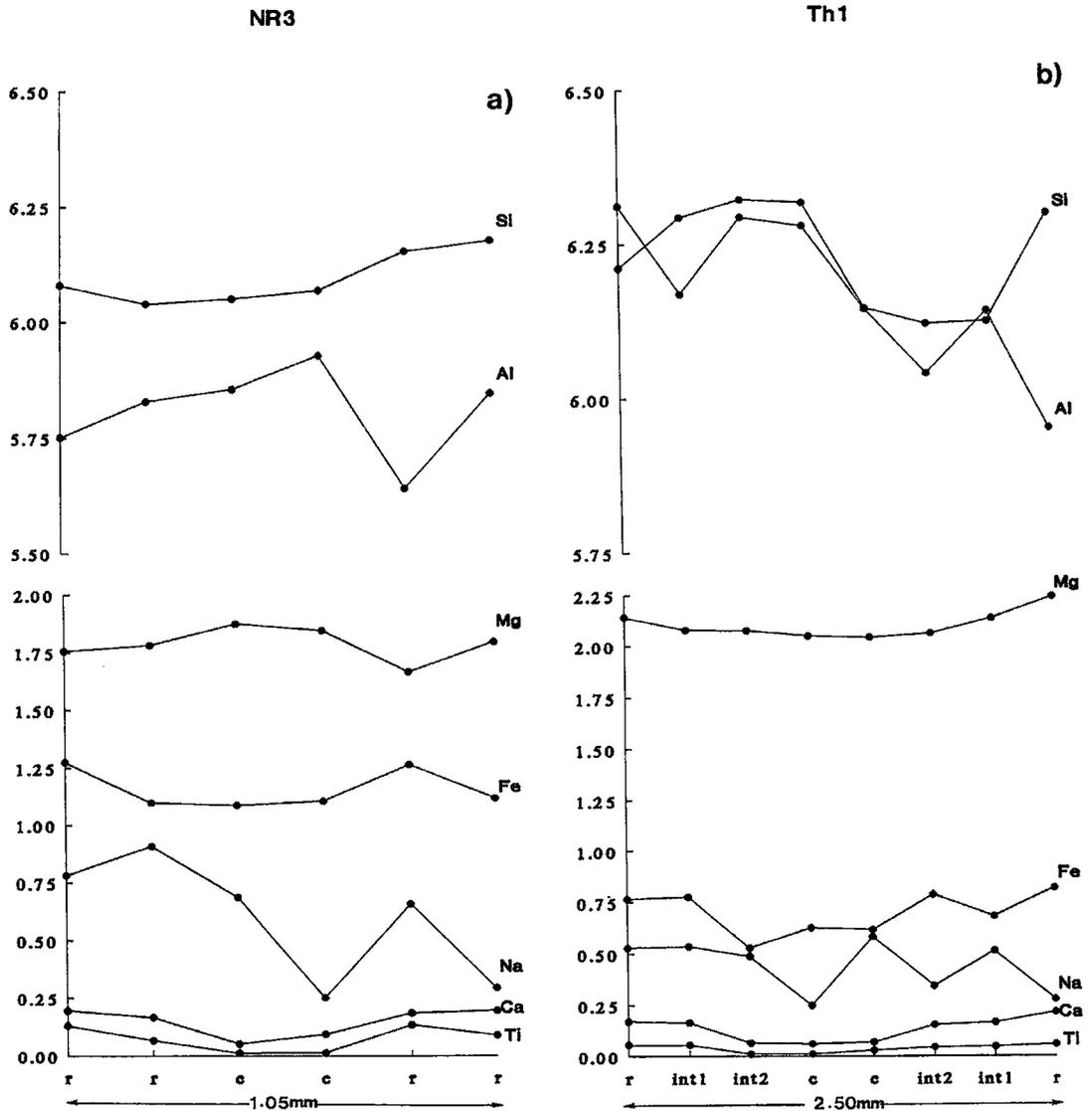


FIG. 4. Rim-to-rim profiles, expressed as cations per formula unit, of representative grains of tourmaline from: a) Nea Roda and b) Thasos areas.

patchy-color domains. As stated by Gallagher (1988), the lack of a consistent pattern of zoning for Na reinforces the view that Na contents may be governed by local charge-balance considerations rather than by bulk chemistry. Zoning profiles across two representative grains are given in Figure 4.

Following Black (1971), Fortey & Cooper (1986) and Slack & Coad (1988), we suggest that the darker coloration of the rim zones (deep green for Thasos and dark brownish green for Nea Roda) is related to the higher Ti concentrations. A small amount of chemical

change would seem to lead to a fairly pronounced optical change.

DISCUSSION AND CONCLUSIONS

The tourmaline crystals occur within quartz veins cross-cutting metamorphosed sedimentary rocks. The origin of the quartz veins could be related to hydrothermal fluids connected to local magmatic activity, to a process of metamorphic remobilization, or to separate epigenetic processes. Field relations

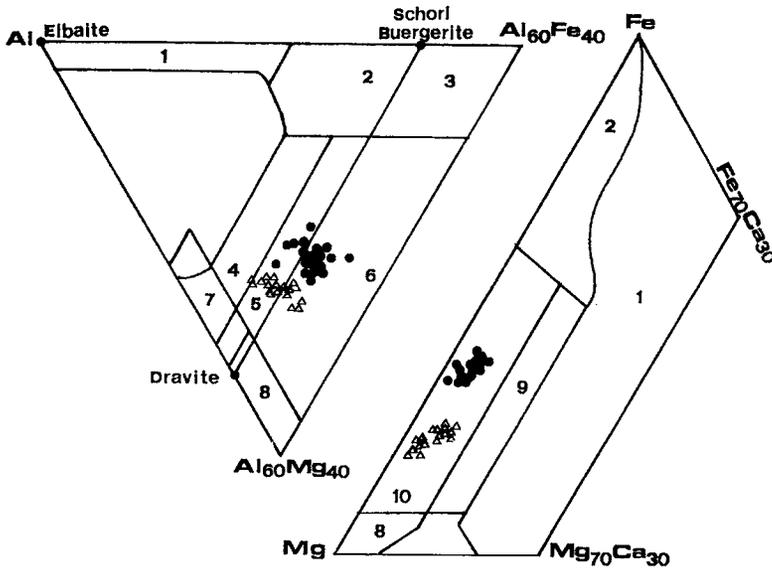


FIG. 5. Triangular Fe–Al–Mg and Fe–Ca–Mg plots showing the tourmaline from Nea Roda and Thasos areas. Symbols as in Fig. 3. Fields as defined by Henry & Guidotti (1985) represent: 1) Li-rich granitic pegmatites and aplites, 2) Li-poor granitic rocks and their associated pegmatites and aplites, 3) Fe³⁺-rich quartz–tourmaline rocks, 4) metapelites and metapsammites coexisting with an Al-saturating phase, 5) metapelites and metapsammites not coexisting with an Al-saturating phase, 6) Fe³⁺-rich quartz–tourmaline rocks, calc-silicate rocks and metapelites, 7) low-Ca meta-ultramafic and Cr-, V-rich metasedimentary rocks, 8) metacarbonates and metapyroxenites, 9) Ca-rich metapelites, and 10) Ca-poor metapelites, metapsammites and quartz–tourmaline rocks.

show that the tourmaline–quartz veins of Nea Roda may be related to nearby felsic plutonic intrusions.

Experimental studies (Werdning & Schreyer 1984) show that tourmaline is stable over a wide range of P–T conditions. Nevertheless, the chemistry of tourmaline may provide information of the environment and conditions of formation. Several investigators have correlated tourmaline chemistry with geochemical environments. As seen in the work of Henry & Guidotti (1985), the major-element compositions of tourmaline usually correlate with the compositions of their host rocks. The Mg–Al–Fe and Mg–Fe–Ca diagrams of Henry & Guidotti (1985) permit a reasonable evaluation of the rock type in which the tourmaline formed. The tourmaline compositions reported here overlap fields 5 and 6 of their classification (Fig. 5), corresponding to the rock types Fe³⁺-rich quartz–tourmaline rocks, calc-silicate rocks, metapelitic and metapsammitic rocks not coexisting with an Al-saturating phase. Consequently, we conclude that the most probable host-rock of the tourmaline is as inferred from Figure 5. This concurs with the field observations, the bulk-rock composition being

migmatitic gneiss (Nea Roda) or high-grade metasedimentary rocks (Thasos).

Several possibilities may be considered for the mineralizing processes: a) the mineralizing fluids were connected with the anatexis of the surrounding migmatized gneisses (*e.g.*, Nea Roda); b) the fluids were magmatic in origin, but during their ascent interacted with the host rock; c) hydrothermal processes post-dating metamorphism were responsible, and d) low-temperature epigenetic or diagenetic processes created the veins and tourmaline in the depositional system of the rocks.

The tourmaline found in most granitic rocks has a composition close to schorl in general (Deer *et al.* 1986, Slack 1982). Results from previous work (Ethier & Campbell 1977, Plimer 1986) suggest that tourmaline from non-evaporitic clastic metasedimentary terranes typically has a composition intermediate between schorl and dravite, similar to those encountered in our study. Thus, we infer that the Mg-contents recorded here preclude the consanguinity of the tourmaline with any granitic rock.

Enrichment in Ga content of the tourmaline suite

also supports a sedimentary origin of their host rocks. Ga is enriched especially in shales, substituting for aluminum in the clay minerals.

The abundance of tourmaline in the quartz veins suggests high concentrations of boron in the hydrous silica-bearing fluids or long-term supply or abrupt change in P-T-X conditions. Boron could be released into circulating fluids from sedimentary precursor rocks by increasing temperature or complexing with other phases. Possible sources of these fluids include the host sedimentary precursor-rocks of the tourmaline-bearing veins, complex through-going hydrothermal solutions, or magmatic or postmagmatic fluids. In any case, we infer that the B-enriched fluids reacted with the host rocks along zones of structural weakness or narrow fractures and through inter- or intragranular pore-space. The reaction involving the B-enriched fluids mixed with permeating meteoric waters and the country rocks resulted in the breakdown of early phyllosilicates and stabilized a tourmaline-bearing assemblage.

Field and textural features show that both the quartz-tourmaline veins and the host metasedimentary rocks have undergone a later tectonometamorphic event. The tourmaline, however, is quite resistant to chemical modification by subsequent metamorphic processes, even under conditions of the granulite facies (Manning & Pichavant 1983), and thus was not affected.

From the present study, we conclude that a) the mineralogy of the tourmaline-bearing veins is limited to quartz for the Nea Roda area, and quartz plus chlorite and sericite for the Thasos area, and b) the fact that the compositional variations of the tourmaline in samples of both areas are relatively minor (except Si, Al and Na) may be explained by coeval nucleation and growth of tourmaline crystals during progressive stages of mineralization.

The Fe/(Fe+Mg) ratio is lower in the Thasos chlorite-associated tourmaline than in the Nea Roda case. The core-to-rim increase of this ratio in both areas is unlike the trends for some other occurrences (Slack & Coad 1988). The zoned crystals of tourmaline show polycyclic variation, with the rim and several intermediate zones changing in many combinations in response to fluid composition.

The core - rim differences in Fe/(Fe+Mg) and Na/(Na+Ca), as well as the variation in Ti, are attributed to the prevailing conditions during the crystallization of tourmaline, the composition of the fluid phase, the rate of crystal growth, *etc.*, and not necessarily to the composition of adjacent mineral phases (Werding & Schreyer 1984).

The overall composition of the tourmaline of Nea Roda and Thasos suggests that these crystals formed in equilibrium with the host-rock schists and gneisses, although small variations imply changing P, T or X conditions for the environment of formation during

crystallization. Field evidence supports development of the quartz-tourmaline veins prior to some deformation and metamorphism, but does not exclude further development. The tourmaline crystals are early and were unaffected compositionally by later metamorphic activity, typical of tourmaline behavior in metamorphic rocks (Henry & Guidotti 1985, Taylor & Slack 1984).

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