

# The application of experimental studies in determining the origin of topaz-quartz-tourmaline rock and tourmaline-quartz rock

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Topaz-quartz-tourmaline rock and tourmaline-quartz rock are unusual rock types which occur in association with granitic rocks in south-west England and elsewhere. A brief review is given of available data concerning the field relationships, mineralogy, chemical composition and fluid inclusion characteristics of examples of both rock types from Cornwall (Roche Rock, Porth Ledden Beach and St Mewan Beacon) and Malaysia, and also for topaz-quartz rock from Australia. Phase equilibria studies which may be applicable in determining the origins of these rock types are also described, together with the results of new experiments which have been carried out to determine their melting relationships. The melting experiments indicate that neither topaz-quartz-tourmaline rock nor tourmaline-quartz rock undergo melting at temperatures up to 800° (2 and above at 1kb pressure, unless a flux is added). The results of experimental studies of F-OH exchange equilibria between coexisting topaz, muscovite and a hydrous fluid phase have been used as a geothermometer for muscovite-bearing topaz-quartz-tourmaline rock from St Mewan Beacon, and indicate an approximate equilibration temperature of 620°C. In the light of the available experimental data, it is considered that the characteristics of natural tourmaline-quartz rock and topaz-quartz-tourmaline rock from Cornwall (and similar rocks from elsewhere) are consistent with a complex hydrothermal origin rather than crystallisation from a volatile-rich residual silicate melt.

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## Introduction

The granites of south-west England are well known for the variety of associated late-stage magmatic and pneumatolytic rock types. Topaz and tourmaline are common components of the late-stage granitic rocks, and are also found in association with hydrothermal mineralisation as a result of greisening or tourmalinisation of host rock adjacent to mineral veins. The presence of these minerals attests to the influence of fluorine and boron in particular at the late-magmatic early hydrothermal stage in the evolution of the batholith.

Two unusual rock types have long attracted the attention of workers in this field. These are tourmaline-quartz rock, known from Roche Rock (St Austell granite) and Porth Ledden Beach (Lands End granite; Charoy 1979) and topaz-quartz-tourmaline rock, known from St Mewan Beacon, St Austell (Collins and Coon 1914). Topaz-quartz-tourmaline rock similar to that of St Mewan Beacon has also been described from peninsular Malaya (Scrivenor 1914). Topaz-quartz rock, with a tourmaline-bearing facies, is known from New South Wales (Eadington and Nashar 1978) and is in many

respects similar to the topaz-quartz-tourmaline rock of St Mewan Beacon. The purpose of this paper is to briefly review the available data relating to these rock types, to discuss their possible origins in the light of the results of recent experimental studies, and to suggest areas for further study.

## Descriptions of the rock types

### *Field relations*

The tourmaline-quartz rock exposed at Roche is in the form of an isolated tot, and contacts between this and adjacent rock types are not exposed. The country rock at surface is killas, but granite has been encountered at no great depth in nearby mines (Dines 1956). Rounded inhomogeneities (possibly xenoliths) are visible in some of the faces of Roche Rock, but are largely inaccessible. Material similar to that of Roche Rock has been found forming the matrix of a breccia of rounded greisen fragments in rubble from mine trials on nearby Belowda Beacon; unfortunately this occurrence is not exposed.

The tourmaline-quartz rock of Porth Ledden Beach is well exposed, and illustrates complex field relationships (Charoy 1979; Pichavant 1979). The tourmaline-quartz rock occurs as subhorizontal irregular planar bodies lying roughly parallel to the granite-killas boundary. It lies below a porphyritic granite facies, and above a tourmaline granite, fragments of which are incorporated as xenoliths. The upper contact is marked by a zone of silicification, which affects both the tourmaline-quartz rock and the porphyritic granite (Pichavant 1979).

The field relationships of the topaz-quartz-tourmaline rock of St Mewan Beacon are recorded by Collins and Coon (1914), who describe the body as a lens occurring at the contact between heavily kaolinised and tourmalinised granite, and tourmalinised killas. Exposure at present is limited to a few small outcrops along the Beacon crest and a small quarry occupied by plant belonging to E.C.L.P. Ltd (localities A and B in Collins and Coon 1914, plate 1). The bulk of the rock is homogeneous, with a clearly visible even distribution of tourmaline grains. Evidence for a change in rock-type as the southern contact is neared is exposed in the quarry, where there is a contact between the topaz-quartz-tourmaline rock and essentially a tourmaline-quartz rock which shows banding parallel to the contact - this may be the contact with 'tourmaline schist' to which Collins and Coon (1914) refer. The dip of the banding is shallow (approximately 40°S) and suggests, if it does indeed lie parallel to the contact, that the contact closely follows the present hill side. Thus the apical parts of the Beacon, north of this exposure, may be not far below the contact which has been removed by erosion. The topaz-quartz-tourmaline rock is itself remarkably hard (it was used to pave grinding pans in china-stone mills - Collins and Coon, 1914) and so is relatively resistant to erosion. On the northern side of the outcrop, close to the site of the contact with granite, lie boulders of topaz, quartz-tourmaline rock cross cut by veins of tourmaline-quartz rock.

The topaz-quartz-tourmaline rock from Gunong Bakau, peninsular Malaya, occurs as sheets cutting now heavily kaolinised biotite granite (Scrivenor 1914). The sheets of topaz-quartz-tourmaline rock are in turn cut by sheets of topaz-bearing aplite, Topaz-quartz rock occurs as dykes and sills cutting, with some brecciation, a roof pendant in the Permian Mole Granite (New South Wales; Eadington and Nashar 1978). This particular example is divided into two facies on the basis of variation in mineralogy (Eadington 1977; described below).

#### *Petrography*

In thin section, both tourmaline-quartz rock and topaz-quartz-tourmaline rock have simple mineralogies and textures. Tourmaline-quartz rock is composed essentially of tourmaline and quartz, with in addition subsidiary apatite noted in the example from Porth Ledden Beach

(Charoy 1979). Tourmaline shows strong colour zoning with repeated alternations of khaki-brown and blue zones, and occurs principally as broken or corroded elongate grains, up to 1-2mm in length. Quartz occurs as grains up to 1-2mm across, with sutured margins, and shows undulose extinction. It is occasionally euhedral when growing into microcavities, in which case optically continuous overgrowths may be observed. When present, apatite is subhedral and clouded with inclusions: it may also be euhedral when growing into microcavities. Secondary tourmaline is also present in material from Porth Ledden Beach, as small blue needles (<50μ long) commonly forming an overgrowth on broken or corroded primary grains. The proportions of tourmaline and quartz vary in hand specimen from material apparently composed entirely of tourmaline to vugs and veins of quartz, and auto-brecciation may be present, as seen at Roche Rock.

The proportions of quartz, topaz and tourmaline in topaz-quartz-tourmaline rock are very variable. Collins and Coon (1914) record that quartz may make UP between 50% and 90% of the rock, whereas both topaz and tourmaline may be almost entirely absent. Where present, topaz makes up to 25% of the rock, as aggregates (up to 5mm across) of many subhedral grains. Tourmaline occurs as subhedral, corroded, elongate grains up to 1-2mm long, which show zoning similar to those in tourmaline-quartz rock. Quartz (up to 1-2mm across) is usually anhedral, with sutured margins and undulose extinction, but is occasionally euhedral when growing into vugs. Muscovite is also present as interstitial books (up to 1mm across) some of which are deformed, and as an alteration product of topaz (which is occasionally completely replaced by muscovite). Accessory minerals include apatite and opaque minerals (which may occur as inclusions in topaz). The bulk of the rock, exposed in the lowermost and central parts of the outcrop, is composed of approximately 60% quartz, 25% topaz and 15% tourmaline. Marginal facies show an increase in the proportions of quartz and tourmaline at the expense of topaz, either individually or together.

According to Scrivenor (1914), the topaz-quartz-tourmaline rock of Gunong Bakau is essentially similar in petrography to that of St Mewan Beacon, but includes accessory cassiterite, arsenopyrite and pyrite. The topaz-quartz rock from New South Wales consists of two facies. The first is a porous (5% by volume) friable rock containing wolframite in addition to topaz and quartz, and is regarded by Eadington (1977) as unaltered. The second, altered, facies is a compact silicic rock with reduced porosity and topaz content, and with accessory biotite, tourmaline, orthoclase, sericite and epidote in addition to wolframite, topaz and quartz.

#### *Chemical composition*

Major element analyses (where available) of both

Table 1. Chemical analyses of tourmaline-quartz rock and topaz-quartz-tourmaline rock.

	1	2	3	4
SiO <sub>2</sub>	71.08	74.48	82.76	81.1
Al <sub>2</sub> O <sub>3</sub>	15.23	12.09	13.16	15.8
Fe <sub>2</sub> O <sub>3</sub>	5.94	6.75	1.35	-
FeO	1.27	0.96	-	0.4
MnO	0.05	0.04	0.04	0.01
MgO	0.26	0.17	0.13	0.17
CaO	0.10	0.25	0.18	0.14
Na <sub>2</sub> O	0.83	0.70	0.07	0.01
K <sub>2</sub> O	0.08	0.07	0.12	0.1
TiO <sub>2</sub>	0.17	0.14	0.01	0.03
P <sub>2</sub> O <sub>5</sub>	0.36	0.46	0.08	n.d.
F	0.43	0.37	3.73	4.6
B <sub>2</sub> O <sub>5</sub>	4.12	3.51	n.d.	n.d.
H <sub>2</sub> O+	1.04	0.82	n.d.	n.d.
total	100.96	100.81	101.45	102.36
O≡F	0.20	0.17	1.57	1.95
total	100.76	100.64	99.88	100.41

1 and 2) tourmaline-quartz rock, Porthledden Beacon (Charoy 1979)

3) topaz-quartz-tourmaline rock, St Mewans Beacon, St Austell (author's analysis) Total Fe as Fe<sub>2</sub>O<sub>3</sub>

4) topaz-quartz rock, New South Wales (Eadington and Nashar 1978) Total Fe as FeO.

n.d. = not determined.

tourmaline-quartz rock and topaz-quartz-tourmaline rock are given in Table 1. The compositions of both rock types will vary greatly with varying proportions of the constituent minerals, and so three of the four analyses given must be considered merely as examples. The analysis given for the topaz-quartz rock from New South Wales is for a homogenised composite of many samples (Eadington and Nashar 1978), and so may be representative of this rock type. The important characteristics of these rock types are that they are low in alkalis and alkaline earth elements in particular, and that the tourmaline-quartz rocks are rich in boron (~4% B<sub>2</sub>O<sub>3</sub>) and the topaz-quartz-tourmaline rocks are rich in fluorine (up to ~ 41/2% F).

#### *Fluid inclusion data*

Relatively few fluid inclusion data are available for tourmaline-quartz rock. Charoy (1979) presents data for tourmaline-quartz rock from Porth Ledden Beach, which includes a maximum observed homogenisation temperature for two-phase inclusions of 480° (2, with most primary inclusions showing homogenisation temperatures between 220-260° and 270-330°C. Some inclusions decrepitated on heating above 480°C, and have homogenisation temperatures greater than this. Fusion temperatures show a wide range, from -3 to -26°C, corresponding to equivalent salinities of 5-25

equiv, wt. % NaCl. The complexity of the fluid inclusion data suggests that the tourmaline-quartz rock of Porth Ledden Beach may have crystallised in equilibrium with solutions of varying salinity, and which may have boiled, at temperatures in excess of approximately 300°C (Charoy 1979). The fluid inclusion assemblage may owe its complexity to successive events rather than a single hydrothermal event.

At present, there have been no fluid inclusion studies of topaz-quartz-tourmaline rock from St Mewan Beacon. However, fluid inclusion data are available for the topaz-quartz rock of New South Wales. Homogenisation temperatures for primary fluid inclusions from this example lie in the range 570-620°C, and the salinity is estimated to be 57 wt. % total salts (Eadington and Nashar 1978). The inclusions are very rich in daughter minerals (as can also be seen in thin sections of material from St Mewan Beacon), and Eadington and Nashar report the presence of K, Na, Ca, Cl, Fe and F, These results suggest that the topaz-quartz rock of New South Wales may have formed at higher temperatures, and in equilibrium with a fluid of higher salinity, than the tourmaline-quartz rock of Porth Ledden Beach.

## Previously proposed origins

Both magmatic and hydrothermal origins have been proposed for tourmaline-quartz rock. The occurrence at Roche has been described as an extreme example of tourmalinisation of a granitic dyke (Ussher and others 1909), but the lack of relic textures or gradation into granitic material led Hatch and others (1972) to the conclusion that the body may have crystallised from a volatile-rich residual magma. Recent studies of both Cornish occurrences led Charoy (1979) to suggest that tourmaline-quartz rock may essentially have crystallised from a hydrothermal solution, which separated from granitic magma enriched in boron by differentiation, at or near to the solidus. A magmatic origin has also been proposed for examples of topaz-quartz-tourmaline rock (Collins and Coon 1914; Scrivenor 1914; Eadington and Nashar 1978) in view of the field relationships and lack of replacement textures. In addition, Eadington and Nashar (1978) report the presence of glass inclusions in topaz from the Australian topaz-quartz rock, and briefly report the results of preliminary melting experiments using this rock type, in which topaz was found as a liquidus phase, with the solidus at 1kb being below 650°C.

## Application of experimental studies

Experimental studies which relate to the origin of tourmaline-quartz rock and topaz-quartz-tourmaline rock may be divided into (a) phase equilibria studies of melting relationships and (b) studies of F-OH exchange equilibria between vapour and silicate phase. Each will be discussed separately.

### *Studies of melting relationships*

Existing studies do not entirely or satisfactorily demonstrate that either tourmaline-quartz rock or topaz-quartz-tourmaline rock may have a simple origin through crystallisation from a melt of similar composition. It is known that the presence of both boron and fluorine causes a marked reduction in the solidus temperature for granite (Chorlton and Martin 1978; Manning 1981), and that the liquidus temperature in the system  $B_2O_3$ - $SiO_2$ - $H_2O$  at 1kb may be as low as 450°C (with 90%  $B_2O_3$ ; Pichavant 1979). Consequently fluorine and boron-rich residual melts may be expected to form at a late-stage in the crystallisation of a granite body, at temperatures of the order of 600°C or less at 1kb (Manning 1981; Chorlton and Martin 1978). Furthermore, Glyuk and Anfilogov (1973), in a study of phase relationships in the system natural granite -  $H_2O$  - HF at 1kb, show a field of quartz, topaz and vapour with 4-7% F in the charge. This field is bounded by a solidus curve at between 670-700°C. Taken at face value, this suggests that the assemblage topaz-quartz begins to melt at temperatures of this magnitude. Experiments of the

type carried out by Glyuk and Anfilogov have been duplicated, and it was found that with the use of hydrofluoric acid the silicate material partially dissolves, even before reaching run temperature, leaving residual material enriched in silica, alumina and fluorine (see Manning 1978). The assemblage topaz-quartz described by Glyuk and Anfilogov (1973) may therefore be residual, representing a non-equilibrium assemblage, and so makes it difficult to interpret and apply the results of this study.

The results of the phase equilibria studies mentioned above cannot be directly applied to determine the origin of tourmaline-quartz rock and topaz-quartz-tourmaline rock. However, they do provide a number of constraints, especially with respect to the lower temperature limit for magmatic processes at 1kb pressure ( $\leq 600^\circ\text{C}$ ). In the absence of directly applicable phase equilibria studies, melting experiments using natural materials may be used to determine the high-temperature high-pressure phase relationships of both rock types. A number of melting experiments using topaz-quartz rock from New South Wales have already been carried out (Eadington and Nashar 1978) and suggested that this rock type could have crystallised from a melt. Unfortunately, precise details of the experimental procedure were not published. In order to assess the likelihood of a magmatic origin for topaz-quartz-tourmaline rock a series of melting experiments have been carried out using material from St Mewan Beacon. In addition, similar experiments were carried out to determine whether tourmaline-quartz rock (from Roche Rock) would melt under similar conditions of temperature and pressure. The experiments were carried out using cold-seal hydrothermal pressure vessels ('rod bombs') run at 1kb pressure and at temperatures between 650°C and 850°C for up to 17 days (Table 2). The solid starting materials (crushed tourmaline-quartz rock and topaz-quartz-tourmaline rock from rubble at Roche Rock and St Mewan Beacon) were encapsulated with 20% water inside platinum capsules. Two capsules were prepared for each experiment, one containing the powdered rock encapsulated with water alone, the other containing powdered rock to which 5% cryolite ( $Na_3AlF_6$ ) had been added as a flux. The results of the experiments are given in Table 2. In neither case was melting detected for samples encapsulated with water alone and run at temperatures up to 800°C (2 or more, but with the addition of cryolite melting was observed for charges produced at temperatures in excess of 750°C. Complete melting was never observed. The results of these experiments suggest that it is most unlikely that either tourmaline-quartz rock or topaz-quartz-tourmaline rock could have crystallised directly from a residual silicate melt.

The observed high salinity of the fluid inclusions suggests that alkalis and their salts may have played an important part in the formation of both tourmaline-quartz rock and topaz-quartz-tourmaline rock. Further experiments need

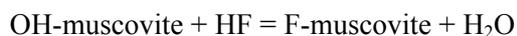
Table 2. Results of melting experiments using tourmaline-quartz rock and topaz-quartz-tourmaline rock. All experiments were carried out at 1kb. In each case, column a is for compositions encapsulated with water alone, column b with 5% added cryolite. A vapour phase was present in all run products, top = topaz; tour = tourmaline; qz = quartz and g = glass

Temperature °C	Time days	tourmaline-quartz rock		topaz-quartz-tourmaline rock	
		a	b	a	b
850	7	tour, qz	g, tour, qz	-	g, top, tour, qz
800	10	-	-	top, tour, qz	g, top, tour, qz
790	14	tour, qz	g, tour, qz	-	-
750	14	tour, qz	trace g, tour, qz	top, tour, qz	trace g, top, tour, qz
710	14	-	tour, qz	-	-
650	17	-	-	-	top, tour, qz

to be carried out to determine the stability of both rock types in the presence of various salts. However, the interpretation of the results of experiments involving a solute-rich aqueous phase would be complicated by the difficulty of quenching the aqueous phase without the condensation of glassy spheres. Thus it may be possible to demonstrate that either rock type may *dissolve in* an aqueous phase of a certain composition at high temperatures, but the presence of quenched glassy spheres may be wrongly interpreted as suggesting that melting of the silicate phase had taken place.

#### Studies of F-OH exchange equilibria

A number of experimental studies have been carried out to determine the F/(F + OH) ratio of certain F-OH bearing minerals in equilibrium with a fluorine-bearing fluid of known composition. Using the results of such studies, the equilibrium constant for reactions such as

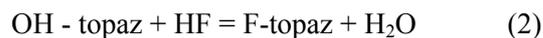


$$K = \frac{f_{\text{H}_2\text{O}}^f \cdot a_{\text{F-muscovite}}^a}{f_{\text{HF}}^f \cdot a_{\text{OH-muscovite}}^a}$$

can be evaluated as a function of temperature. Once this is known, the fugacity ratio for HF and H<sub>2</sub>O in a fluid coexisting with a given F-OH phase of known composition can be calculated for a desired temperature. If two different F-OH phases coexist, and are both assumed to be in equilibrium with the same fluid phase, then we can solve two simultaneous equations to determine the equilibrium temperature. The equilibrium constants for the muscovite and topaz F-OH exchange reactions have been determined as functions of temperature at 2kb pressure (Munoz and Ludington 1977; Rosenberg 1978 respectively), and are as follows:



$$\log K_1 = \frac{2100}{T \text{ (°K)}} - 0.11$$



$$\log K_2 = \frac{-6338}{T \text{ (°K)}} + 11.204$$

If we assume that the topaz-quartz-tourmaline rock of St Mewan Beacon formed by hydrothermal processes, and that the interstitial books of muscovite are primary (especially as some books show signs of deformation before complete consolidation) then equations (1) and (2) can be solved to give the temperature of equilibration and fluid composition at the time of equilibration. In order to do this, the compositions of muscovite and topaz from St Mewan Beacon have been determined by electron microprobe analysis, and are given in Table 3. Solution of these equations gives the temperature of equilibrium as 620°C ± 40°C, and the fluid composition as 0.04% HF ± 0.003% HF (0.02M ± 0.0015M HF). The uncertainties given arise from consideration of the precision limits for the determination of fluorine in both the natural minerals, and the synthetic minerals used to derive the equilibrium constant-temperature relationships. The figures given also rely on the assumptions that ideal mixing of F and OH takes place within the silicate phases and (as H<sub>2</sub>O and HF) in the aqueous phase, that the aqueous phase is a supercritical fluid, and that there is no major change in the equilibrium temperature with pressure. These assumptions clearly do not hold, especially as the aqueous phase is a complex mixture of salt and water. The geothermometer uses the ratio of H<sub>2</sub>O and HF fugacities in the aqueous phase, which will be little altered by the presence of small quantities of dissolved salts. However, the effect of large quantities of dissolved salts on the H<sub>2</sub>O and HF fugacities is unknown

without further experiments, and may affect the calculated equilibrium temperature. This limitation in particular should be borne in mind, but in the absence of further information, the topaz muscovite geothermometer indicates that the topaz-quartz-tourmaline rock of St Mewan Beacon may have crystallised in equilibrium with a hydrothermal fluid at approximately 620°C.

Table 3. Chemical compositions of muscovite and topaz from topaz-quartz-tourmaline rock (St Mewan Beacon). Atomic proportions have been calculated assuming complete occupation of the F-OH site; H<sub>2</sub>O contents were calculated from the atomic proportion of OH. Total iron is recorded as Fe<sub>2</sub>O<sub>3</sub>.

	Muscovite		Topaz	
SiO <sub>2</sub>	47.25		33.93	
Al <sub>2</sub> O <sub>3</sub>	36.77		55.31	
Fe <sub>2</sub> O <sub>3</sub>	0.28		-	
K <sub>2</sub> O	10.47		-	
F	0.59		17.38	
H <sub>2</sub> O	4.26		1.69	
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O = F	0.25		7.32	
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Total	99.37		100.99	
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Si	6.243	8.000	Si	1.024
Al	1.757		Al	1.968
Al	3.970	3.998	F	1.659
Fe	0.028		OH	0.341
K	1.765			
OH	3.753	4.000		
F	0.247			

## Conclusions

The available data, derived from petrographic, fluid inclusion and experimental studies, are insufficient to determine precisely the origin of either tourmaline-quartz rock or topaz-quartz-tourmaline rock. However, certain constraints may be deduced. The field relations of both rock types suggest that they formed from a medium which was able to behave intrusively causing brecciation of the host rock. A significant hydrothermal component is indicated by the reported presence of rugs and the high porosity of certain examples of both rock types. Melting experiments so far carried out suggest that both tourmaline-quartz rock and topaz-quartz-tourmaline rock are highly refractory, as are their constituent minerals. Taken together, the data indicate that neither tourmaline-quartz nor topaz-quartz-tourmaline rock (or topaz-quartz rock as in the New South Wales example) are likely to have a simple magmatic origin from the crystallisation of residual granitic melts of peculiar composition. In view of the fluid inclusion data in

particular, it is considered that these rock types were formed by hydrothermal processes which may have been multistage and complex and may have commenced soon after the close of magmatic crystallisation. Further studies are required, in particular to compare the topaz-quartz-tourmaline rock of St Mewan Beacon with the topaz-quartz rock of New South Wales, and to obtain fluid inclusion data for the St Mewan Beacon rock. Further experimental studies of the stability of both rock types in salt solutions at high temperatures and pressures are also desirable and should be carried out in conjunction with fluid inclusion studies.

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