Some topaz-, sillimanite-, and kyanite-bearing rocks from Tanzania

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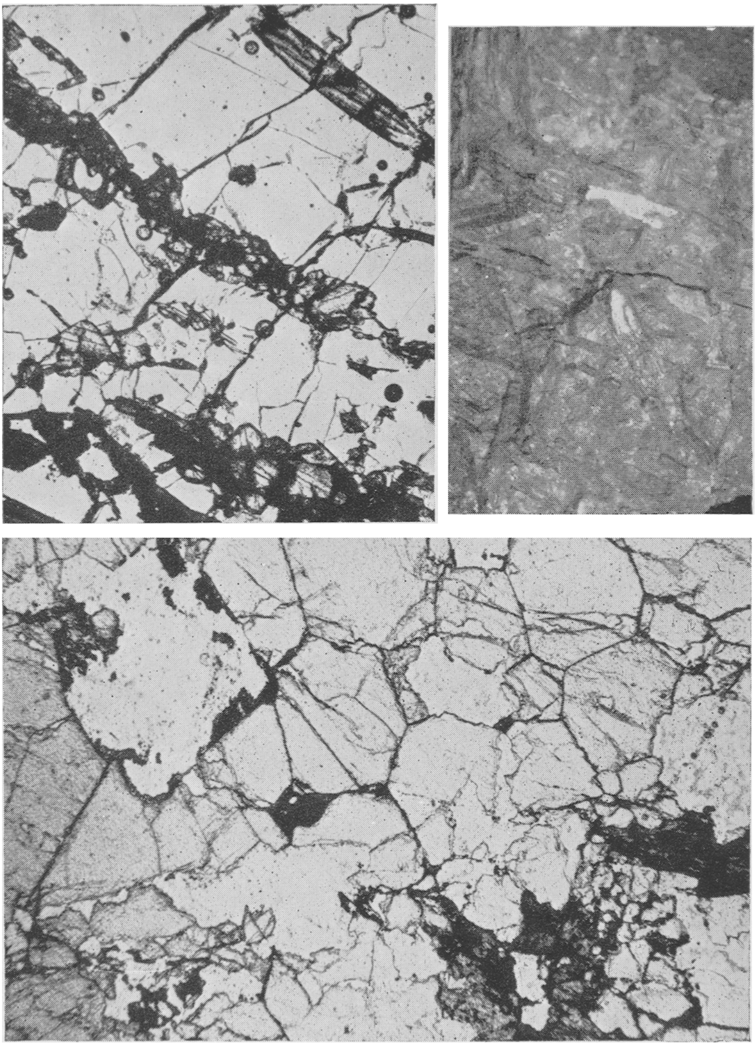
Summary. Some sillimanite–kyanite quartzites, which may grade into sillimanite–kyanite topazfels, from NE. Tanzania are described. In view of the suggestion that a solid-solution series might exist between topaz and sillimanite, the cell dimensions and some optical properties of both minerals were investigated. No abnormal results, suggesting solid solution, were obtained and the coexistence of topaz, sillimanite, and kyanite is ascribed to particular physico-chemical conditions.

SILLIMANITE-BEARING rocks are common in the Pre-Cambrian rocks of the Usagaran (? ≡ Mozambiquian) system of the Usambara Mountains, Tanga province, NE. Tanzania. Bagnall, Dundas, and Hartley (1965) describe them briefly as graphite–sillimanite schists, and kyanite and sillimanite schists and gneisses, associated with granulites, almandine–amphibolites, and other rocks of high metamorphic grade. The rocks are strongly foliated (foliation striking 350°, dipping 20° E.) and lineated (lineation striking 52°, plunging 18°).

A group of sillimanite and kyanite quartzites, heavily stained with iron oxides, at Kwemahambalawe, 10 miles north-north-west of Lushoto, has been found to include topaz-bearing rocks. Also in this vicinity, bands of relatively pure kyanite are found in which the kyanite occurs as aggregates of large radiating crystals forming boulders ('boulder rosettes') in a schistose matrix of felted sillimanite and smaller blades of kyanite (Bagnall *et al.*, 1965, p. 16).

The Kwemahambalawe quartzites comprise at least four rock types. Firstly, type (A) is an iron-stained schistose quartzite containing small acicular prisms of sillimanite with minute bubble inclusions of low refractive index, elongated parallel to the *c* axis, a little kyanite, and accessory rutile and zircon, the latter always in well-rounded detrital grains. In this rock the sillimanite and other heavy minerals constitute about 10 % of the whole, quartz making up the remainder (fig. 1).

The second type, (B), is a coarsely schistose iron-stained quartz–



FIGS. 1–3: FIG. 1 (*top left*). Photomicrograph of rock type (A), showing prisms and cross-sections of sillimanite, with some iron ore and accessories, in quartz. Plain light, ×30. FIG. 2 (*top right*). Fractured surface of rock type (B), showing large prisms of sillimanite (dark and, totally reflecting, white). Natural size. FIG. 3 (*bottom*). Photomicrograph of rock type (C). Mainly topaz, with (*bottom right*) kyanite; (*top left*) broken cross-section of sillimanite; (*left lower centre*) quartz. Plain light, ×16.

sillimanite rock, in the approximate proportions: quartz 85 %, sillimanite 14 %, and kyanite, with a little topaz, rutile, and zircon, 1 %. In this rock the sillimanite occurs as coarse needles up to ½ in. or more in length, with a generally parallel lineation in the plane of the schistosity (fig. 2).

The third and most interesting type, (C), is essentially an iron-stained topazfels, consisting of a saccharoidal mosaic of topaz crystals (*circa* 75 %), with about 15 % of sillimanite, 5 % of quartz, and 5 % of kyanite, rutile, and zircon (fig. 3). The sillimanite is clearly visible in hand specimen as coarse needles, again up to ½ in. in length, in no apparent orientation.

Lastly, the fourth type, (D), is a quartzite, less iron-stained than the remainder, with sparsely distributed, very pale yellow, ovoid topazes in sub-parallel orientation and a few grains of rutile and zircon. In this rock sillimanite appears to be absent and the topaz and accessories represent only some 1 % of the total, the remainder being quartz.

Experimental results and discussion

Rocks transitional from one zone to another, in which kyanite and sillimanite coexist, are known to be fairly widespread (see Deer, Howie, and Zussman, 1962, p. 126; Hodge, 1966); that the assemblage should locally include topaz is, however, unusual, although a similar paragenesis is known from Murka Hill, near Taveta, Kenya, about 100 miles to the NNW. (du Bois, 1966). The common paragenesis of topaz is in acid igneous rocks, generally associated with late-stage pneumatolytic action, but its presence in a regional metamorphic terrain is ascribed to regional metasomatism of fluorine (Deer *et al.*, 1962, p. 149).

The triple assemblage topaz–sillimanite–kyanite is the more surprising in the light of Coes's (1955) experimental work. He synthesized, amongst other high-pressure minerals, the three Al_2Si0_5 polymorphs and topaz, the optimum temperatures and pressures being 700° C and 10 000 atmospheres (andalusite and sillimanite) and 900° C and 20 000 atmospheres (kyanite and topaz). Coes noted that kyanite occurred as a by-product in the synthesis of the two other polymorphs but not of topaz; the simultaneous crystallization of kyanite and topaz was never observed and Coes comments that the formation of one seems to exclude formation of the other. Coes also noted that sillimanite never forms in the absence of fluorine and considers it possible that an extensive or even complete series of solid solutions exists between sillimanite and topaz. It has also been suggested by other workers that structural water might be present in one or more of the Al_2SiO_5 polymorphs or, in the case of sillimanite, structural fluorine.

In view of the different structures of the two minerals it is difficult to see how complete solid solution could exist between them. Both are orthorhombic but, if they are orientated so that the cell parameters *a*, *b*, and *c* are to have similar values, they are found to have different space groups (topaz, *Pmnb*; sillimanite, *Pbnm*). If they are differently orientated to have the common space group, *Pbnm*, the cell-edge lengths *a* and *c* are widely different. Assuming some solid solution relationship to exist, therefore, the most likely possibility seems to be the presence of limited solid solution near each end member. At intermediate compositions or lower temperatures or both, a solvus might operate and, as in the alkali feldspars and pyroxenes, some form of exsolution relationship might result.

If any type of solid solution is present, it might be indicated by abnormal cell dimensions in either or both of the two phases, resulting from distortion of the lattice of the host member. Their coexistence in the topazfels from Tanzania seemed to the writer to present an opportunity of testing the hypothesis on natural rocks. The cell dimensions of topaz from two rocks and sillimanite from three were accordingly determined by a least-squares method from powder photographs taken on an 11·46-cm diameter Philips camera, using Cu-$K\alpha$ radiation; in the case of rock type (B) determinations were made on two sillimanite fractions that appeared during separation to show a slight density variation. These dimensions, together with those determined for a sillimanite from the Mogok ruby mine, Burma, and a topaz from a loose boulder by the Cairngorm granite, Scotland, which were used as standards, are given in table I. It is considered that there is no significant variation in the dimensions; moreover, consideration of the reflection intensities of the powder patterns showed no variation that could be considered systematic and therefore significant.

The α refractive index of the two minerals and $2V_\gamma$ of the topaz from types (B), (C), and (D) and the sillimanite from types (A) and (C) were determined and are given in table I. The values for topaz suggest an OH/(OH+F) molecular percentage of about 18 or 19, or near the centre of the range (Deer *et al.*, 1962, p. 148). In no case were any significant departures from the values given in Deer *et al.* (1962) encountered.

From the evidence given above there seems to be little support for the theory that intermediate members of a solid-solution series commonly exist in nature. Coes (1955) considers that, whilst it crystallizes in the pressure–temperature range of kyanite, the formation of sillimanite (and andalusite) is controlled by chemical factors, in contrast to kyanite, which is insensitive to the chemical composition of the reaction mixture. It would seem that the formation of topaz is similarly controlled by chemical factors.

TABLE I. Cell-dimensions and some optical data for topaz and sillimanite

	Topaz			
Rock type	(B)$_2$*	(C)	(D)	Cairngorm granite
a	—	4·638 Å	4·644	4·639
b	—	8·810	8·779	8·795
c ($\pm$0·005)	—	8·366	8·369	8·369
α ($\pm$0·002)	1·618	1·618	1·619	1·616
$2V_\gamma$ ($\pm$1°)	61°	60°	58°	—

	Sillimanite				
Rock type	(A)	(B)$_1$*	(B)$_2$*	(C)	Mogok, Burma (B.M. 1920, 17)
a	7·462	7·468	7·453	7·472	7·477
b	7·677	7·660	7·659	7·667	7·655
c ($\pm$0·005)	5·767	5·763	5·762	5·763	5·757
α ($\pm$0·002)	1·660	1·660	1·659	1·660	1·659
$2V_\gamma$ ($\pm$1°)	28°	—	—	25°	—

Space groups: *Pbnm* Radiation: Cu-$K\alpha$ (Ni filtered)

* Rock type (B): 1 = light fraction; 2 = heavy fraction.

However, if the materials used by Coes (1955) to synthesize the aluminium silicates are considered, no obvious chemical explanation for the presence of topaz is apparent: for sillimanite, kaolin + various fluorides; for kyanite, kaolin + $Al(OH)_3$; for topaz, $AlF_3 + Al(OH)_3 + SiO_2$. Other workers have synthesized topaz from the thermal hydrolysis of AlF_3 and SiO_2 at 750° to 950° C (Schober and Thilo, 1940); and from Na_2SiF_6, amorphous Al_2O_3, and H_2O, heated to 500° C at 4000 bars pressure (Michel-Lévy and Wyart, 1946). Both Bien and De Keyser (1962) and Löcsei (1963) have found topaz to be a by-product in the synthesis of mullite from the thermal interaction of AlF_3 and kaolinite; the first-named authors note that it is only present at temperatures between 795° and 1000° C. It might also be mentioned that Grubenmann (1910, p. 165) proposed that the richness in alumina of some Swiss sillimanite-gneisses might be explained by the introduction of aluminium fluoride, giving the reaction: $2AlF_3 + 3H_2O = Al_2O_3 + 6HF$.

Commenting on this suggestion, Harker (1950, p. 250) considered that if alumina could be introduced in this way, the resulting product would be topaz rather than sillimanite.

If Coes's reaction mixture is postulated as occurring in nature, the following reaction may be proposed:

$$2AlF_3+24Al(OH)_3+13SiO_2= \underset{\text{topaz}}{3Al_2SiO_4F_2}+\underset{\substack{\text{sillimanite}\\\text{(or kyanite)}}}{10Al_2SiO_5}+\underset{\text{water}}{36H_2O}$$

If the system is anhydrous, the substitution of $12Al_2O_3$ for $24Al(OH)_3$ will result in the disappearance of water from the products. The proportion of topaz to sillimanite plus kyanite of 3:10 given above is chosen to balance the equation; since the reactants in all probability would vary considerably in different parageneses, this ratio is highly variable, as is the proportion of sillimanite to kyanite. And since Coes obtained only topaz from his reaction mixture, some catalytic or further condition must exist in nature if the reaction is to take place.

Another possibility must be considered: the conversion of sillimanite (or kyanite) into topaz, according to some reaction based on: $Al_2SiO_5+F_2= Al_2SiO_4F_2+O$ at temperatures below those at which both sillimanite (1545° C) and topaz (850°–900° C) are known to break down into mullite plus other phases.

No satisfactory explanation for the assemblage topaz–sillimanite–kyanite, reconcilable with the experimental work of Coes, is apparent and it is suggested in conclusion that, whilst the presence of some fluorine is necessary for the formation of sillimanite, the formation of topaz requires the presence of a higher concentration of fluorine, probably with precise and as yet undefined physico-chemical conditions. Whether the process should be regarded as metasomatic in the sense of implying pneumatolysis is debatable. The fluorine necessary for the formation of sillimanite is accepted as part of a normal metamorphic closed system and it is preferred here to regard topaz, like sillimanite and kyanite, as a primary mineral resulting from the high-grade regional metamorphism of impure (aluminous) siliceous sediments in an environment locally having a relatively high fluorine content. It is possible that close examination might reveal the presence of small quantities of topaz coexisting with sillimanite (and kyanite) in similar regionally metamorphosed rocks.

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