

A Model of Aluminium Silicate Accretion in Metamorphic Rocks*

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Abstract. Textures resulting from the replacement of biotite by kyanite in biotite-rich schists of the polymetamorphic lower Basement Complex of the Luangwa Bridge area, Zambia, are described. This mode of kyanite growth, and several examples of Al_2SiO_5 growth by recrystallisation of cordierite, are used as a basis for a model of aluminium silicate accretion in rocks which have undergone polymetamorphism or polyphase metamorphism. Starting from the assumption that Al_2SiO_5 phase transformations are usually accomplished by a fluid intergranular phase it is suggested that aluminium concentration in the fluid, in addition to other factors, has a profound effect on the final assemblage of aluminium silicates.

Introduction

Compared to some other rock-forming minerals, the study of the Al_2SiO_5 minerals has developed along definite lines. The aluminium silicates became more widely studied at a time of great advances in experimental mineralogy and their treatment became rather one-sided. Thus the present knowledge of the modes of formation of naturally-occurring Al_2SiO_5 minerals, and of the factors controlling their coexistence, in spite of recent efforts, does not compare favourably with the amount of information and sophistication reached in the experimental and theoretical study of the Al_2O_3 - SiO_2 system.

It is realised that some associations and time relations of the Al_2SiO_5 polymorphs in individual metamorphic terrains are of ubiquitous occurrence while others are quite rare. This state reflects the existence of a predominating repetitive trend in the development of fold belts. Obviously, the frequency of occurrence of individual patterns of coexistence of the Al_2SiO_5 polymorphs bears little relation to their significance for understanding the factors controlling the coexistence of the aluminium silicates.

Chinner (1966), in his discussion of the Al_2SiO_5 system, concluded that the natural occurrence of the polymorphs will not be affected by the composition of the intergranular fluid. This conclusion is relevant only to assemblages of minerals of strictly coeval crystallisation. The general textural relationships, however, indicate that in the crystallisation of metamorphic rocks we are dealing rather with a sequence of reactions than with a strictly coeval crystallisation of all the

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phases. At present, we have no example of the undisputed coeval crystallisation of kyanite, andalusite, and sillimanite or even of any two of the polymorphs. On the contrary, there is abundant evidence of the natural sequential formation of the polymorphs (Hietanen, 1956; Chinner, 1966, and others).

Kyanite Growth by Replacement of Biotite

A supracrustal sequence of gneisses, migmatites, and quartzites in the lower Basement Complex of the Luangwa Bridge area (Central and Eastern Provinces, Zambia) shows structural and mineralogical evidence of superposition of orogenic belts and polymetamorphism (Vrána, in prep.). The early metamorphic history of the complex was associated with folding on north-west trending axes, tentatively correlated with the Ubendian orogeny (2150–1650 m.y.). This was a fully developed cycle of deformation, regional metamorphism and granitisation. During deformation at a considerably later date the rocks of the complex were refolded along north-east trending axes. This refolding and refoliation, which may be either Lufilian or Irumide, strongly modified pre-existing small-scale structure and was associated with a relatively high-pressure amphibolite facies recrystallisation.

Microscopic study of specimens from this area has shown that some biotite-rich portions of the migmatites, especially biotite schlieren grading to schistose biotitite, and biotite mica schists, were affected by a remarkable alumina metasomatism during the superimposed metamorphism (Fig. 1a, b). In a group of 27 specimens of mica schists and related rock types, 7 contain newly-formed kyanite produced by alumina metasomatism. Some of the resulting textures, and the relationship between the pre-existing sillimanite and newly-formed kyanite are described below.

The new kyanite has grown by the internal replacement of biotite, with the development of unusual crystal shapes (Fig. 2a) or of fine-grained aggregates (Fig. 2b). There is also a well-pronounced tendency for kyanite to be idioblastic against biotite and xenoblastic against felsic minerals (Fig. 2a).

Commonly, the kyanite contains inclusion trails of opaque minerals arranged parallel to the (001) planes of the pre-existing biotite. The replacement by kyanite of biotite originally intergrown with sillimanite has resulted in swarms of sillimanite needles becoming enclosed in kyanite metacrysts (Fig. 4a) and careful microscopic examination shows that the sillimanite remains intact.

In nearly all thin sections showing kyanite as a replacement of biotite, the former is accompanied by accessory to trace amounts of a strongly pleochroic tourmaline of greyish-blue to greenish-blue colour parallel to ω ($\omega \cong 1.653$). This type of tourmaline is confined to rocks showing metasomatic growth of kyanite and is optically different from tourmalines in other rock types. This association of metasomatic kyanite and tourmaline presents an interesting analogy with that of such "pneumatolytic" minerals as coarse muscovite, tourmaline and topaz, with metasomatic sillimanite.

Kyanite growth by replacement of biotite resulted from a restricted to pervasive metasomatism (see Fig. 1a). The volumes affected range from narrow biotite bands in migmatites, which are only a few millimetres thick, to bands of biotite

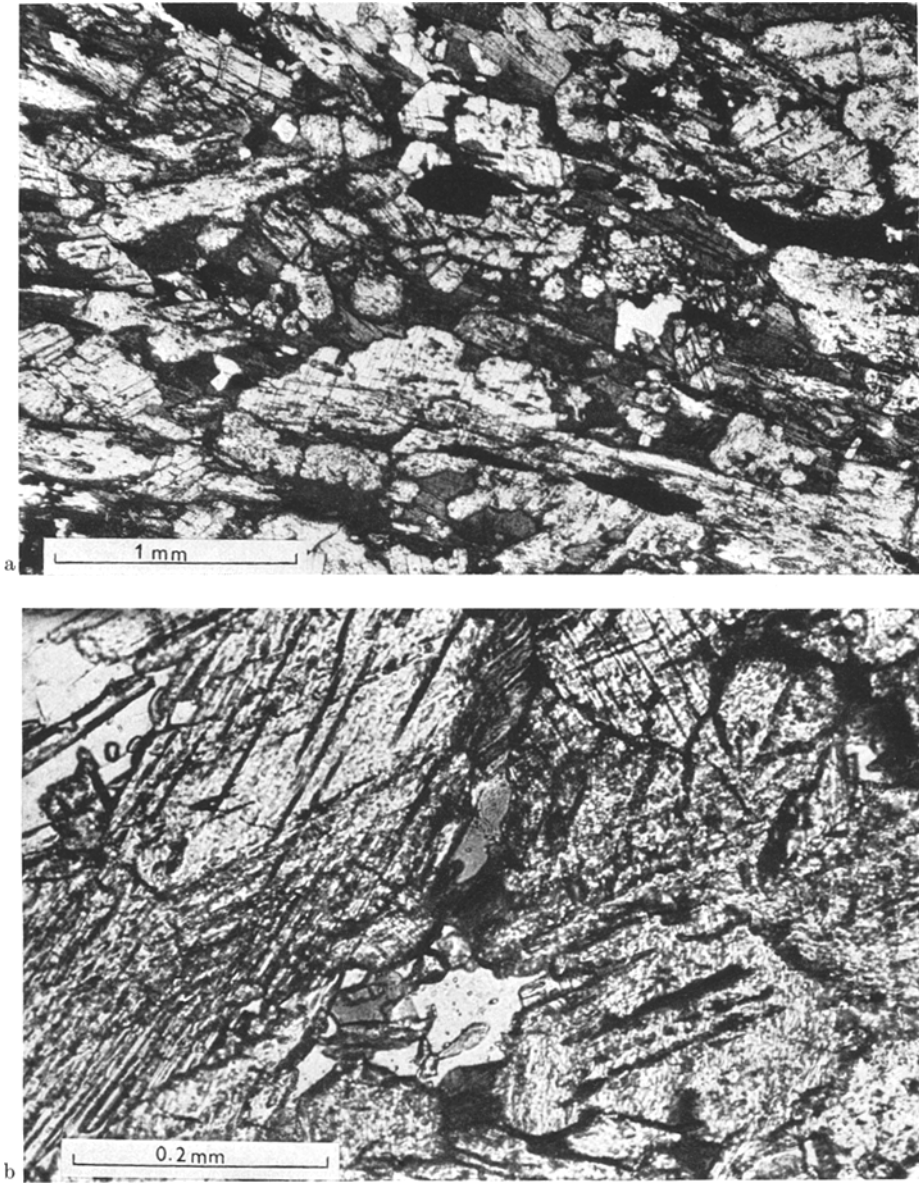


Fig. 1. a Nearly monomineralic biotitite containing accessory sillimanite has been extensively replaced by kyanite. The kyanite is of metasomatic origin and shows inclusion trails of opaque minerals derived from the replaced biotite (see Fig. 1b at higher magnification). Specimen 9GC* 258 of schistose biotitite schlieren in a polymetamorphic migmatite; Tondo River. (* Specimens and thin sections are included in the collections of the Geological Survey Department, Lusaka, Zambia.) b Inclusion trails of opaque minerals running continuously from bottom left to top right through two kyanite grains of different orientations. The inclusion trails were formed during the replacement of biotite by kyanite. A formerly biotite-rich band in a polymetamorphic banded migmatite; Mupwasha River; specimen 9GC 132

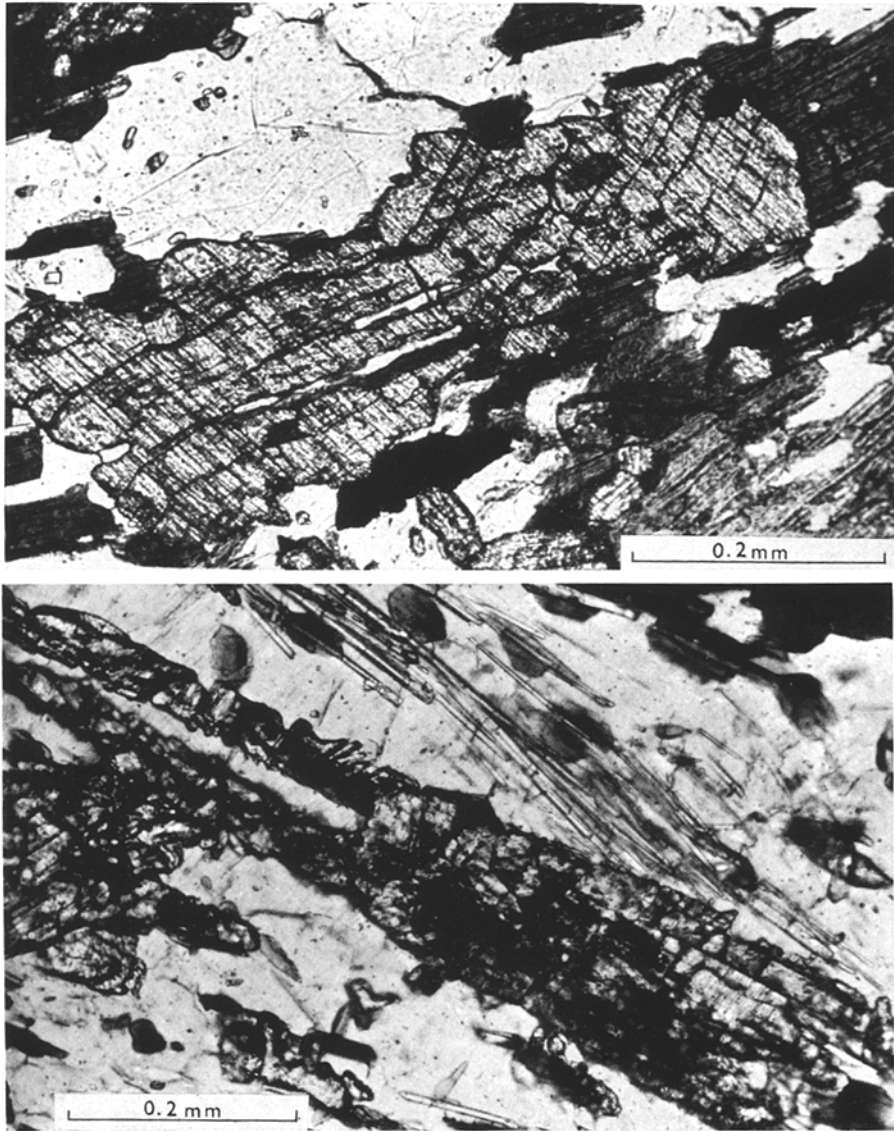


Fig. 2. a A single grain of kyanite replacing biotite. The kyanite grain, sectioned nearly parallel to (010), shows an unusual morphology with an $a:c$ ratio of about 4:1. Note the idioblastic shape of the kyanite against biotite at the upper right corner. A polymetamorphic quartz-kyanite-biotite-sillimanite-garnet schist; Mwateshi River; specimen 9GC 282. b Fine-grained kyanite aggregate replacing and pseudomorphing biotite; earlier fibrolitic sillimanite is preserved intact. Banded migmatite; Mupwasha River; specimen 9GC 132

mica schist several metres in thickness. Possibly balance was attained through a material exchange with the much larger volumes of surrounding rock-types in which no kyanite was produced.

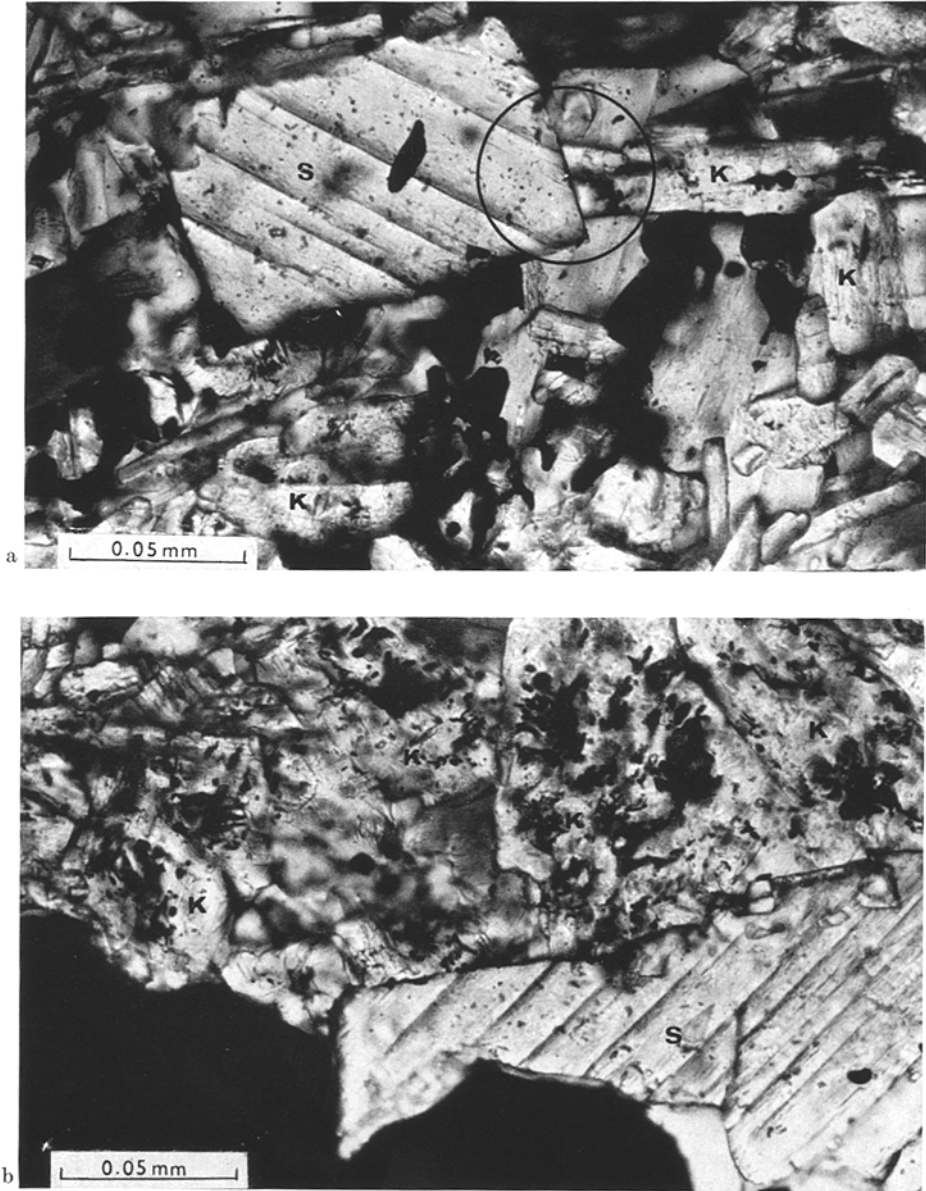


Fig. 3. a A single crystal of sillimanite (*S*) surrounded by newly-formed prismatic kyanite (*K*), some relict biotite and relatively abundant opaque minerals. Note the character of the sillimanite-kyanite boundary (encircled). A polymetamorphic quartz-antiperthite-biotite-kyanite-garnet-sillimanite mica schist; Confluence of the Luangwa and Kapila rivers; specimen 9GC 332. b Skeletal sillimanite (*S*) and a large grain of opaque mineral (bottom left) overgrown by newly-formed kyanite (*K*). The kyanite was formed by replacement of biotite and contains numerous inclusions of opaque mineral (the central and upper parts of the figure). Specimen 9GC 332

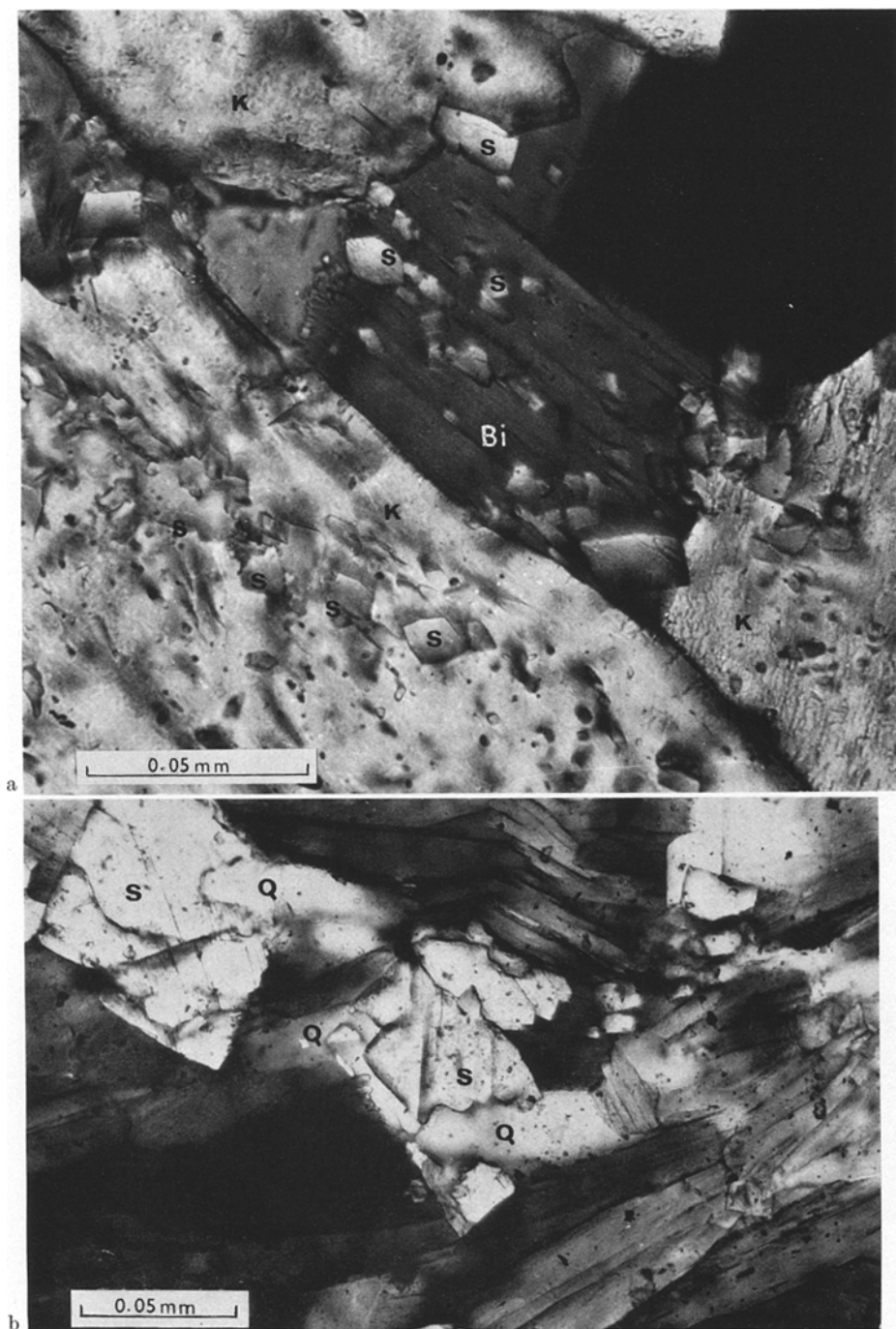
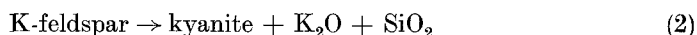
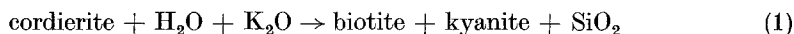


Fig. 4. a Biotite (*Bi*) carrying fibrolitic sillimanite, which occurs mainly as cross sections. The surrounding kyanite (*K*), which has grown by replacement of biotite, inherits some of the sillimanite. Sillimanite inclusions are especially abundant in the lower left of the figure. Several individual sillimanite crystals are partly enclosed in biotite and partly in kyanite. Schistose biotite schlieren in a polymetamorphic migmatite; Tondo River; specimen 9CG 258. b Sillimanite (*S*), corroded and partly replaced by quartz (*Q*). Note the post-crystallisation deformation of biotite. Specimen 9GC 332.

In the Luangwa Bridge area a variety of metamorphic reactions were involved in the crystallisation of kyanite. Besides growth by replacement of biotite, some kyanite was produced during recrystallisation of cordierite and by associated dealkalisation of potash feldspar. This diversity in the modes of kyanite formation suggests that the pressure and temperature conditions were well within the kyanite stability field.

Kyanite Growth by Recrystallisation of Cordierite

The polymetamorphic gneisses and migmatites of the Luangwa Bridge area may contain pseudomorphs up to a centimetre long of acicular kyanite and biotite or phlogopite. Where the newly-formed minerals are fine-grained the pseudomorphs are macroscopically comparable to pinitic pseudomorphs after cordierite. Indeed, a gneiss carrying pseudomorphs of this type has been found in close proximity to cordierite gedritite containing fresh cordierite. These pseudomorphs are interpreted as having been formed by the recrystallisation of pre-existing cordierite. Potash feldspar grains occurring near the pseudomorphs are mutually separated by thin composite veinlets consisting of quartz and fine-grained acicular kyanite. It is suggested that these veinlets were formed by the de-alkalisation of potash feldspar during which the potassium migrated towards the cordierite sites and into the newly-formed biotite. The textures are thus interpreted in terms of two simultaneous and complementary reactions as follows:



These pseudomorphs are in certain respects similar to the kyanite-andalusite-chlorite pseudomorphs after cordierite described by Lal (1969). However, in the present case no andalusite was observed, and the place of chlorite is taken by biotite or phlogopite.

An identical type of cordierite recrystallisation has recently been recognised in the polymetamorphic cordierite-sillimanite granulites of the Chipata District (Vavřda and Vrána, in press). In these rocks recrystallisation of cordierite to pseudomorphs consisting of kyanite, biotite (phlogopite), garnet, with or without (?) sapphirine, usually resulted in the presence of two aluminium silicates that is, the granular (coarse prismatic) sillimanite and the newly-formed kyanite. In some specimens the total content of aluminium silicates is probably higher than at any earlier paragenetic stage. The relative abundance of kyanite-bearing pseudomorphs in the Chipata District is due not only to the former ubiquitous presence of cordierite in the granulites but also to the regional extent of the recrystallisation which involved rocks now cropping out over hundreds of square kilometres (Vavřda and Vrána, in press). The recrystallisation of cordierite to kyanite-bearing pseudomorphs apparently did not involve a whole-rock metasomatism. Nevertheless, during the period of recrystallisation the aluminium potential was apparently high. The formation of kyanite according to the previously mentioned reactions (1) and (2) presents another example of aluminium silicate accretion. In a few specimens of polymetamorphic granulite the coarse, granulitic sillimanite is inverted to kyanite. This inversion may have resulted from the activity of a

neutral pore fluid at a date later than the recrystallisation of cordierite to kyanite-bearing pseudomorphs.

Andalusite Growth by Recrystallisation of Cordierite

A specimen of nodular sillimanite-cordierite gneiss within the contact zone of the younger Ntimba Granite (Chipata District, Eastern Province, Zambia) shows recrystallisation of its early cordierite as andalusite-biotite pseudomorphs (Vavřda and Vrána, in press). The potassium required for the crystallisation of the biotite was derived by de-alkalisation of the potash feldspar, and tiny prismatic grains of andalusite are now found along the potash feldspar grain boundaries. There does not appear to have been large scale metasomatism involving a change of the original whole rock composition. The alumina necessary for the accretive growth of andalusite was made available by the recrystallisation of cordierite. In the same rock concentrations of early sillimanite in the form of nodular aggregates survived the period of andalusite growth.

Discussion

From an examination of the literature it might appear that some of the problems relating to the occurrences of the aluminium silicates have many almost insoluble facets. Turner's statement (1968, p. 77) illustrates this apparent complexity: "The coexistence of andalusite, sillimanite and kyanite in some pelitic schists, and perhaps the association of cummingtonite and anthophyllite in magnesium hornfels, possibly reflects the metastable condition of one or more of the phases concerned. It is likely then that kyanite, sillimanite or andalusite may be a metastable member of the metamorphic assemblage even when occurring alone. The well known tendency for sillimanite to replace mica suggests that crystallisation of this particular phase rather than andalusite or kyanite may be due in some instances to a smaller value ΔS in the breakdown of mica to sillimanite as contrasted with andalusite or kyanite. Sillimanite could then be metastable from the moment of its formation".

This statement certainly expresses a pessimistic view. Turner (1968), however, quotes evidence indicating a more regular and tractable behaviour of the Al_2SiO_5 polymorphs in which kyanite is developed mostly in the moderate- to high-pressure kyanite zone of regional metamorphism, andalusite in the lower temperature regimes of low-pressure regional metamorphism or of contact metamorphism and sillimanite supersedes both kyanite and andalusite in the realm of high temperatures. Miyashiro (1949) noted the existence of this general pattern and Chinner (1966) concluded that in the majority of natural occurrences the Al_2SiO_5 polymorphs do conform to the pattern established by experimental studies.

Contrary to the widespread belief that the Al_2SiO_5 polymorphs have an inherent tendency to metastable persistence it is suggested that the phenomena are dependent upon the composition of the intergranular fluid at the time of growth of the later polymorph.

It can be assumed that Al_2SiO_5 phase transformations are usually accomplished by solution and precipitation by an active intergranular fluid. Where the fluids

are oversaturated in alumina, the existing Al_2SiO_5 polymorph persists. This can be postulated from the fact that an intergranular fluid oversaturated in alumina cannot dissolve a pre-existing Al_2SiO_5 mineral; it will stabilise the existing polymorph and probably cause the further crystallisation of an aluminium silicate, possibly of another species. It can be further postulated that in most cases the identity of the existing polymorph will be immaterial. The composition of the fluid is clearly of predominant importance and it is only in some relatively rare cases of limited range of composition and of P - T conditions that some other physico-chemical factors, such as differences in the free energy, solubility, nucleation and reaction rates, etc., are important.

Thermodynamic calculations and experiments show rather small energy differences among individual polymorphs (Richardson *et al.*, 1969). This, of course, suggests that metastable persistence and metastable growth should be common. On the other hand, cases of inversion of one polymorph to another are well known and examples are described by Hietanen (1956), Pitcher (1965), Chinner (1966), Hollister (1969), etc. They undoubtedly demonstrate that even the small ΔG may be sufficient to obtain inversion under natural conditions.

A qualitative compositional classification may be made as follows:

1. Fluid undersaturated in aluminium which can dissolve and eliminate a pre-existing aluminium silicate, provided there is a relatively high diffusion rate, as in the case of silicification associated with a superimposed metamorphism. An example of the resultant intermediate texture is given in Fig. 4 b.

2. Fluid of a "relatively neutral composition" which facilitates transformation of one polymorph into another. If the transformation can take place only between narrow limits of fluid composition, the aluminium silicate assemblages formed will be dependent on the time during which the composition favourable to transformation is maintained. Cases of partial inversion, as described by Hietanen (1956), Chinner (1966, Fig. 4A), Fyfe (1969, pl. 1) and others, also are probably dependent on the time during which the intergranular fluid maintains a certain composition. Occasionally a change in P - T conditions while the transformation is under way can be an additional factor.

3. Fluid oversaturated in aluminium, producing metasomatic growth of an aluminium silicate. Examples include the commonly described later crystallisation of sillimanite in kyanite- or andalusite-bearing rocks (Chinner, 1961), and some of the cases of aluminium silicate accretion described in this paper.

Several inferences can be drawn:

1. Disregard of the controlling role of the composition of the intergranular fluid has resulted in erroneous concepts of the metamorphic conditions prevailing during the evolution of rocks carrying two or three Al_2SiO_5 polymorphs. For example Hietanen (1956) believes that the three aluminium silicate polymorphs do have their respective stability fields, albeit overlapping to some extent. Considering temperature and pressure as the sole controlling factors she arrives at the conclusion that during metamorphic evolution, the P - T conditions were hovering around the Al_2SiO_5 triple point. If the controlling role of the aluminium concentration in the intergranular fluid is taken into account, however, it is possible that during some of the stages of crystallisation of the Al_2SiO_5 polymorphs

the pressure-temperature conditions might have been very remote from the triple point.

In general it can be concluded that an association of two or three Al_2SiO_5 polymorphs does not imply proximity of the pressure-temperature parameters to an univariant line or to the triple point, unless it can be proved that the crystallisation of the polymorphs was coeval.

2. Where two or three polymorphs formed during subsequent periods of metamorphism, they represent a record of the physical conditions during those periods as indicated by the species and succession of the aluminium silicates.

3. The examples of kyanite growth by replacement of biotite are also of interest in considering the origin of fibrolitic sillimanite, which is known to be formed by a similar mechanism (Pitcher, 1965). However, the fibrolitic sillimanite does not—and cannot—preserve the relict texture of the host biotite.

Kyanite and andalusite are usually formed relatively early, during progressive metamorphism of unaltered pelitic sediments and present little textural indication of the reactions by which they were formed. Sillimanite usually forms at higher temperatures after the rock is fully recrystallised and textures arise indicative of its growth by replacement of a pre-existing mineral or by reaction among some of the pre-existing minerals (besides inversion of an earlier Al_2SiO_5 polymorph). The textural features of this sillimanite cannot be considered evidence for metastable growth of sillimanite or fibrolite as believed by some petrologists.

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