THE AMERICAN MINERALOGIST, VOL. 53, MAY-JUNE, 1968

ON THE RELATIVE SCARCITY OF PARAGONITE

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ABSTRACT

Consideration of low- to medium-grade paragonite-bearing rocks suggests that the presence of this mineral is related to relative enrichment in Al_2O_3 and/or $NaAlO_2$. Rocks with highly aluminous minerals, such as chloritoid, kyanite, etc., commonly have paragonite; whereas less aluminous rocks which contain biotite rarely have paragonite. It appears that rocks with insufficient Al_2O_3 to plot above the garnet-chlorite join on an AKFm projection will not develop paragonite.

As metamorphic grade increases joins such as garnet-chlorite or staurolite-chlorite will break and bring in the assemblage alumino-silicate+biotite. If these reactions occur before the upper stability limit of paragonite is exceeded, then paragonite can occur in alumino-silicate+biotite-bearing assemblages. Present data indicates that kyanite is the only alumino-silicate to occur in this assemblage. Hence it is postulated that the pT curve for the breakdown of paragonite and those for the reactions producing alumino-silicate +biotite must cross so that paragonite is unstable before the assemblage andalusite (or sillimanite)+biotite becomes stable.

INTRODUCTION

The absence of paragonite in the pelitic schists of northwest Maine, despite a range of metamorphic grades, has led the writer to consider the controls on the occurrence of this mineral.

There is a growing literature on the field occurrence (Rosenfeld, 1956; Rosenfeld, *et al.*, 1958; Albee, 1965A; Zen, 1960; Zen *et al.*, 1964; and Zen and Albee, 1964) and on the experimental relations (Eugster and Yoder, 1954A, 1954B, 1955; and Iiyama, 1964) of this mineral. Nonetheless paragonite seems to be uncommon and to occur in restricted assemblages.

Paragonite is seldom found to be coexisting with andalusite, pyrophyllite, or biotite (nor with albite, according to Tröger, 1967, p. 506). Sillimanite is not reported with paragonite, but kyanite is common with it although in many cases in quartz-kyanite veins (e.g., Zen *et al.*, 1964) or kyanite-quartzite (Espenshade and Potter, 1960), in which biotite (and plagioclase) are usually absent. Nonetheless the assemblage kyanite+biotite+staurolite+paragonite is recorded in the Alps (see #39 and 40, Table 1, Zen and Albee, 1964). Andalusite (or pyrophyllite)+biotite +paragonite is not found and may be impossible as an equilibrium assemblage. It is evident from Zen and Albee (1964, Table 1) that paragonite is usually associated with muscovite and commonly with chloritoid and chlorite. In summary, paragonite seems to be uncommon in shcists and to be more restricted in associations than muscovite or biotite. It is most common in medium grades of metamorphism. Zen and Albee

(1964, p. 921) state, "Everything else being equal, rocks in the almandine and low kyanite zones of metamorphism might be the most likely hosts to paragonite."

Some of the apparent scarcity of paragonite is due to the difficulty in identifying it. In this paper other explanations which involve both compositional controls and stability relations will be proposed to explain this paucity and the restricted associations. It is well to note that many ideas suggested below probably apply to pyrophyllite also.

IDEALIZED COMPOSITIONAL CONTROLS ON THE OCCURRENCE OF PARAGONITE AND THE NA-CONTENT OF MUSCOVITE

An idealized model of the expected phase relations of paragonite can best be portrayed on a series of AKNa projections (Thompson, 1961). Figures 1A-C are examples of the expected phase relations for paragonite from low to high grades of metamorphism. There is little doubt that relative enrichment of $NaAlO_2$ or Al_2O_3 can produce paragonite-bearing assemblages as long as paragonite is thermodynamically stable. Moreover it is apparent that even the paragonite content of muscovite in the assemblage muscovite+plagioclase will be related to the relative $NaAlO_2$ and/or Al_2O_3 content of a given rock. Possibly the reported occurrences of paragonite+glaucophane (Harder, 1956) in rocks of low Al_2O_3 are the result of high $NaAlO_2$. The relationship between relative Al_2O_3 content and the mole percent of paragonite in muscovite is well shown by comparing muscovite in the assemblage muscovite+plagioclase to that in the assemblage muscovite+plagioclase+sillimanite (see Fig. 1C). Evans and Guidotti (1966) found that muscovite in the first assemblage contains 6–10 mole percent paragonite whereas that in the second assemblage has 14 mole percent paragonite. One specimen with the assemblage Ksp+plagioclase+muscovite has only 4 mole percent paragonite in the muscovite. A further illustration of the close relation between the Al_2O_3 content (as implied by the assemblage present) and mole percent of paragonite in muscovite has been observed by the writer in the upper staurolite grade schists in the Rangeley area, Maine. Basal spacings of the muscovite in these rocks indicate sodic muscovite (in assemblage X_3 but close to X_2 in Fig. 1B) in rocks with staurolite present. Rocks with chlorite +biotite+garnet have a more K-rich muscovite and those with only biotite have the most K-rich.

Clearly the geometry of Figures 1A–C and the examples cited above leave little doubt that the relative enrichment of $NaAlO_2$ and/or Al_2O_3 influences the presence or absence of paragonite and also controls the mole percent of paragonite dissolved in muscovite in paragonite-free rocks.

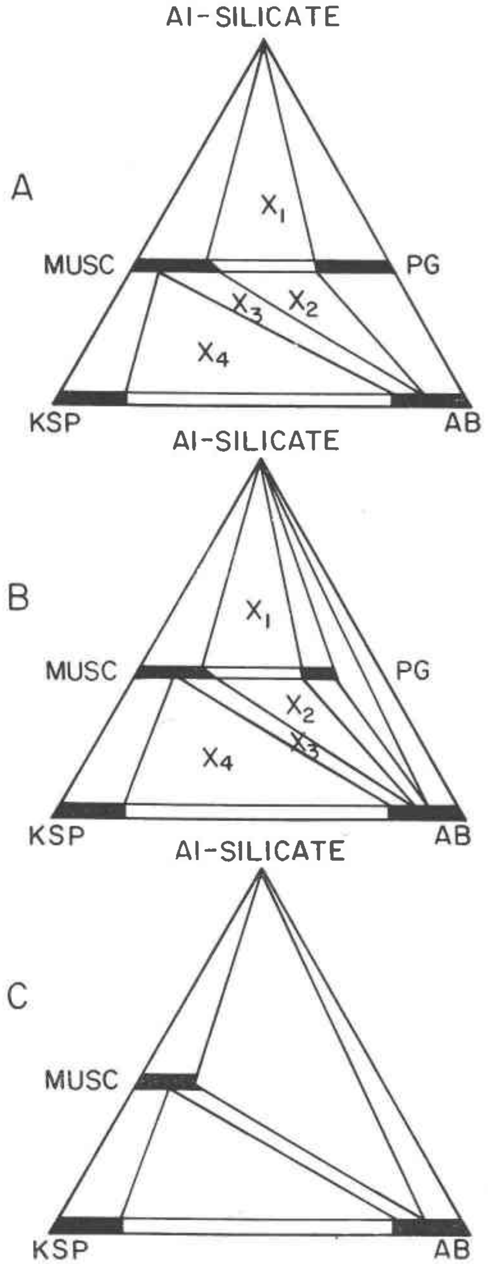

FIG. 1. Schematic diagrams for the phase relations in the system K_2O-NA_2O-Al_2O_3-SiO_2 (Excess) just below and above the stability limit of paragonite, after Thompson (1961).

OBSERVATIONS ON THE RELATION BETWEEN NATURAL ASSEMBLAGES AND ROCK COMPOSITION

The phase relations in pelitic schists can be described by KNaAl and KAlFm projections (Thompson, 1957; 1961). CaO is the only significant component not included, but it is not important for the ideas expressed below.

Figures 1A and B illustrate some common phase relations in the system KNaAl at medium grades. Assemblage X_1 is common with kyanite as the aluminosilicate. Eugster and Yoder (1954a, p. 113) mention pyrophyllite in X_1 instead of kyanite. However, documentation of pyrophyllite with paragonite is not convincing as the pair is not recorded in the detailed reports (Albee, 1957; Cady *et al.*, 1962) of the two areas cited by Eugster and Yoder. Andalusite in X_1 is recorded by Harder (1956) but is very rare. Moreover, from Harder's descriptions it is not certain if equilibrium assemblages were present. Clearly X_1 requires very Al-rich bulk compositions (*e.g.*, kyanite-quartzite).

Assemblages X_2 and X_4 have also been found but X_3 is by far the most common in the system. This concentration in X_3 is notable as it suggests a limited range in bulk composition for most pelites. Inasmuch as $KAlO_2$ is usually considerably more abundant than $NaAlO_2$ in pelitic schists it is clear that variation in Al_2O_3 will be most important in determining whether paragonite will be present or absent. Hence variation of assemblage from X_4 to X_3 to X_2 is in most instances going to be the result of increased Al_2O_3 rather than $NaAlO_2$.

Except for the phase at the Al-corner, no change occurs in Figures 1A or B during metamorphism until paragonite breaks down, giving Figure 1C.

Figures 2 and 3 are representative KAlFm projections for other phase relations at medium grades. Paragonite can not be represented on a KAlFm projection. However, observations show that only those assemblages which lie in the high Al_2O_3 portions of the KAlFm projections are likely to have paragonite present. For example, the data of Harder (1956), Tröger (1967), Zen and Albee (1964), and Hoschek (1967) indicate paragonite in assemblages containing minerals such as chloritoid, kyanite, chlorite, staurolite, etc. In contrast, low Al_2O_3 assemblages, such as those with biotite, rarely have paragonite. In general it appears that (with few exceptions) bulk compositions which plot below a line connecting garnet and chlorite do not contain paragonite.

Figure 3 (after Fig. 5 of Albee, 1965) is highly instructive for illustrating the relation between composition and paragonite paragenesis. The assemblages recorded on that diagram (in order of decreasing Al_2O_3) are kyanite+chlorite+chloritoid, garnet+chloritoid+chlorite, garnet+

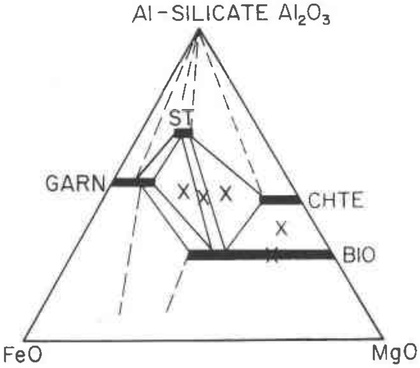

FIG. 2. Schematic projection from muscovite of the mineral assemblages in the upper staurolite zone. Quartz in excess based upon the assemblages in the upper staurolite grade, Rangeley Quadrangle, Maine. Guidotti (1967) X = recorded assemblage.

chlorite, and garnet+chlorite+biotite. The first two have paragonite, the third paragonite+albite, and the fourth has only albite. It is evident that the Al_2O_3 content affects the presence or absence of paragonite.

As pointed out by Albee (1952) and emphasized by him in 1965B, an average pelitic schist plots below a line connecting garnet and chlorite on Figure 2. Hence medium-grade schists do not commonly have an aluminosilicate or pyrophyllite because Al_2O_3 is deficient inasmuch as the orientation of tie lines (*e.g.*, garnet-chlorite, staurolite-chlorite, etc.) in the KAlFm system precludes their occurrence in rocks of average Al_2O_3 content (*e.g.*, in Albee, 1965A, kyanite and biotite are incompatible; see Fig. 3). With regard to paragonite, the observations cited above (exemplified

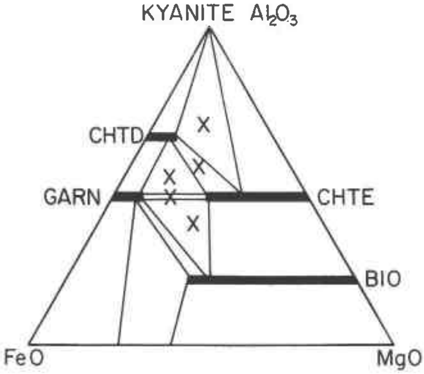

FIG. 3. Schematic representation of the mineral assemblages in the kyanite zone, Lincoln Mtn. quadrangle, Vermont. X = assemblages referred to in text. After Albee (1965), Fig. 5 p. 270.

clearly by the sequence of assemblages in Fig. 3) and the fact that most pelites plot below the garnet-chlorite join indicate that paragonite is rare because the relative Al_2O_3 content of most schists is low and thus sufficient only to produce X_3 on Figures 1A or B rather than X_1 or X_2. Thus the rarity of the pair biotite+paragonite in medium grades is explained; bulk compositions on a KAlFm diagram low enough in Al_2O_3 to contain biotite are very likely to be too poor in Al_2O_3 to permit paragonite (in the KNaAl system). Similarly, but less rigorously so, the pair albite+paragonite is uncommon. Inasmuch as the common bulk compositions which will yield biotite and/or albite are too poor in Al_2O_3 to contain paragonite or aluminosilicates at medium grades, pairs such as andalusite+paragonite, pyrophyllite+paragonite, etc. should be very rare, except in very highly aluminous rocks in which case biotite will be absent.

Hoschek (1967) has demonstrated that relative Al_2O_3 content has an analogous effect on the presence or absence of chloritoid. Indeed the line connecting garnet and chlorite seems to have a somewhat similar role in the paragenesis of chloritoid inasmuch as Hoschek (1967, p. 128) notes that biotite is rare in chloritoid-bearing rocks. It is obvious from Table 1 of Zen and Albee (1964) as well as from the assemblages listed in Hoschek (1967) that chloritoid and paragonite often occur together. Hence it is very likely that the compositional arguments so well documented for the paragenesis of chloritoid by Hoschek (1967) apply equally to the paragenesis of paragonite.

Harder (1956) has also clearly emphasized the importance of high Al_2O_3 in order to produce paragonite-bearing schists. Moreover, although Harder lists several specimens with biotite plus paragonite (without an aluminosilicate) and even a few with paragonite in the assemblage garnet+biotite+chlorite, he nonetheless emphasizes the general incompatibility of biotite and paragonite. Harder (1956) states (p. 261) "Ebenso fehlt Paragonite sobald durch das Auftreten von grossen Mengen Biotit der Aluminium-wert herabgesetzt und der Magnesium- bzw. Eisengehalt heraufgesetzt wird." Possibly paragonite in the assemblage biotite+garnet+chlorite results from a higher than normal content of $NaAlO_2$. The possibility of high $NaAlO_2$ producing paragonite in a low alumina rock is evident from Figure 1.

Thus the scarcity of paragonite and some of the limitations on its associations (*e.g.*, biotite+paragonite) are a function of composition in conjunction with the orientation of tie lines in the KAlFm and KNaAl systems.

METAMORPHIC CONTROLS ON THE PARAGENESIS OF PARAGONITE

The assemblage kyanite+staurolite+biotite+muscovite+paragonite +quartz suggests that metamorphic controls may in some cases enable rocks with lower Al_2O_3 (as low as the phase region for biotite on a KAlFm projection) to contain paragonite+biotite. Consideration of the PT controls of paragonite must of course be viewed in conjunction with compositional controls.

The paragonite stability curve (Eugster and Yoder, 1954a)—even with the modifications suggested below—passes through the andalusite and possibly even the sillimanite pT fields as determined by Newton (1966) and Althaus (1967) (see Fig. 4), and it extends well into the sillimanite field of Holm and Kleppa (1966). The field of paragonite certainly overlaps those of biotite and pyrophyllite. For example, Kerrick (1967) has recently determined that pyrophyllite breaks down to andalusite+quartz $+H_2O$ at $410° \pm 10°C$ at 1.8 kb P_{H_2O}. In the discussion of the preceding section the pairs kyanite+biotite and paragonite+biotite were incompatible. It is evident that in the assemblage kyanite+biotite+paragonite a tie line such as staurolite+chlorite has reoriented to aluminosilicate+biotite by a reaction such as given by Guidotti (1967).

$$\text{staurolite} + \text{Mg-chlorite} + \text{sodic-muscovite} \rightleftharpoons \text{Al-silicate} + \text{biotite} + \text{K-richer muscovite} + \text{Ab} + \text{qtz} + H_2O \quad (1)$$

The topology of Figure 1A or 1B must remain unchanged. The modal amounts of muscovite and chlorite decrease and one can think of the Al_2O_3 thereby released as being moved into the KNaAl subsystem (forming aluminosilicate). Thus a rock initially too poor in Al_2O_3 to have paragonite (X_3) may, as a result of increased grade effecting reactions in the KAlFm system, receive an influx of Al_2O_3 into the KNaAl system, thus producing the assemblage kyanite+paragonite+muscovite (plus biotite).

The pair aluminosilicate+paragonite without biotite could occur in a similar manner if the rock were somewhat higher than normal in Al_2O_3 (somewhat above the garnet+chlorite join) and if generalized reactions such as (2–5) were possible and occur before the garnet+chlorite tie line is broken.

$$\text{staurolite} + \text{qtz} \rightleftharpoons \text{kyanite} + \text{garnet} + H_2O \quad (2)$$

$$\text{staurolite} + \text{qtz} \rightleftharpoons \text{kyanite} + \text{garnet} + \text{chlorite} + H_2O \quad (3)$$

$$\text{chlorite} + \text{qtz} \rightleftharpoons \text{kyanite} + \text{garnet} + H_2O \quad (4)$$

$$\text{chlorite} + \text{chloritoid} + \text{qtz} \rightleftharpoons \text{kyanite} + \text{garnet} + H_2O \quad (5)$$

Many of these are hypothetical reactions and would occur only in rocks of higher than normal Al_2O_3. Moreover, in the writer's experience the garnet+chlorite tie line seems to be broken quite early, thereby making some of these reactions impossible.

The pair biotite+paragonite could also occur before the incoming of an aluminosilicate in rocks with bulk compositions lying just above the garnet+chlorite join by means of reactions such as from Albee (1965B, Table 2):

$$\text{garnet} + \text{chlorite} + \text{musc} \rightleftharpoons \text{staur} + \text{bio} + \text{qtz} + H_2O \quad (6)$$

$$\text{garnet} + \text{chlorite} + \text{musc} \rightleftharpoons \text{chloritoid} + \text{bio} + \text{qtz} + H_2O \quad (7)$$

However, such assemblages with paragonite are not common. This is primarily the result of most bulk compositions plotting below the garnet-chlorite join—such rocks will probably still lack paragonite even after reactions (6) and (7). Moreover it is also possible that (6) and (7) are very close in t and p to those reactions which first bring in the pair aluminum silicate+biotite (*e.g.*, reaction #1) or (from Albee, 1965B, Table 2):

$$\text{garnet} + \text{chlorite} + \text{musc} \rightleftharpoons \text{kyanite} + \text{bio} + \text{qtz} + H_2O \quad (8)$$

$$\text{chlorite} + \text{chloritoid} + \text{musc} \rightleftharpoons \text{kyanite} + \text{bio} + \text{qtz} + H_2O \quad (9)$$

The calculated slopes (Albee, 1965B) for most of these reactions are steep.

An explanation is required for the association kyanite+biotite+paragonite but absence of andalusite (or pyrophyllite)+biotite+paragonite. Consistent with this observation is the suggestion that the PT curves for the reactions which first bring in kyanite+biotite cross the stability limit curve of paragonite as shown in Figure 4.

Excluding reactions involving cordierite (which has not been recorded with paragonite anyway) reactions yielding kyanite+biotite probably all occur at roughly similar T and p. Albee (1965B) has shown that all of these reactions have very steep slopes at 2.5 kb (P_{H_2O}). Reaction 1 is probably important in nature and has recently been defined in detail (Guidotti, 1967); hence it is used below. The analogous reaction of Albee (1965B) has a slope of 78 bars/degree when kyanite is a product, and it can be calculated, using Albee's data and the molar volumes and entropies of andalusite and sillimanite (Clark, 1966), that slopes of 41 and 46 bars/degree result when andalusite and sillimanite are the respective products. Reaction 1 is more complex than Albee's equation, but there is little reason to suspect a pT slope different from that of Albee.

Evans (1965) has determined the pT curve for the reaction:

$$\text{muscovite} + \text{qtz} \rightleftharpoons \text{Ksp} + \text{Al-sil} + H_2O \quad (10)$$

The slope of this reaction at 2–3 kb (P_{H_2O}) is 25, 28, and 33 bars/degree when andalusite, sillimanite and kyanite are the respective products. There is little reason to feel that the reaction

$$\text{paragonite} + \text{qtz} \rightleftharpoons \text{Ab} + \text{Al-sil} + H_2O \quad (11)$$

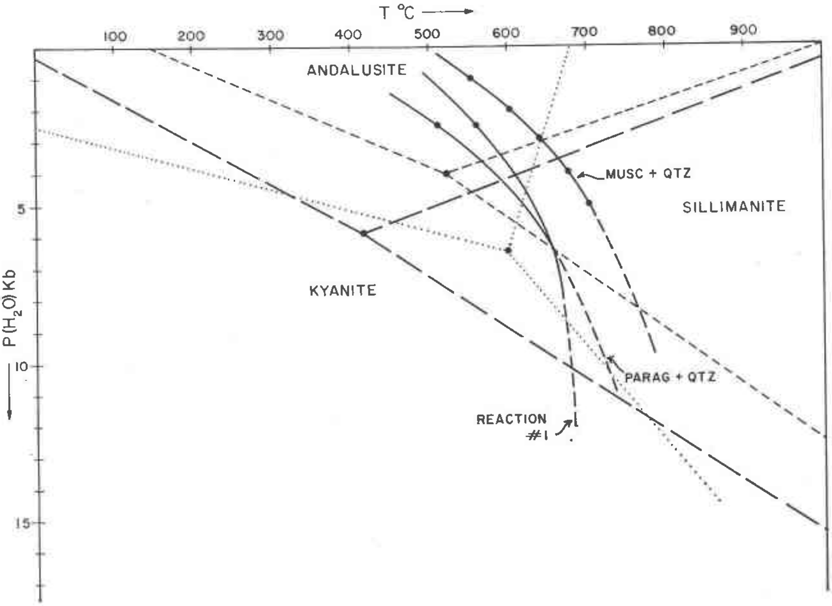

FIG. 4. Schematic relation of the paragonite+qtz curve to the curve for reaction 1 (see text). Muscovite curve from Evans (1965). Dashed lines = Al-Silicate curves of Holm and Kleppa (1966). Small dashes = Al-silicate curve from Newton (1966). Dotted lines = Al-silicate curve from Althaus (1967).

or the geologically more realistic reaction [which occurs at a T slightly above (11)]

$$\text{(Na, K) paragonite} + \text{qtz} \rightleftharpoons \text{Ab} + \text{Al-sil} + \text{(K, Na) musc} + H_2O \quad (12)$$

will have a significantly different slope from (10). Hence the slopes of (1) and (11) or (12) could cross as shown on Figure 4. The slope suggested for (11) or (12) is not very different from that of the experimental curve for pure paragonite (Eugster and Yoder, 1954A, p. 113).

The assemblage data given suggest that the curves on Figure 4 will be close to each other. Geological evidence from the Rangeley area, Maine, also supports the suggestions that paragonite is unstable before the assemblage andalusite+biotite becomes stable. In Guidotti (1966, Fig. 2 and 3) data on the basal spacing of muscovite in pelitic schists clearly suggests that the muscovite in these rocks becomes Na-richer down grade to the upper staurolite zone. Recent additional data from the lower sillimanite, upper staurolite, lower staurolite, and garnet zones shows that the most sodic muscovite occurs in the upper staurolite zone (d(002) = 9.926 Å). At higher and lower grades it is K-richer. From the shape of the muscovite-paragonite phase diagram (Eugster and Yoder, 1955) it is evident that the upper staurolite zone rocks in the Rangeley area must have formed at a temperature close to that for the upper stability limit of paragonite. Reaction (1) initiates the lower sillimanite zone (and the decrease in Na in the muscovite)[1] but would be virtually the same with andalusite as a product. This reaction (producing andalusite) must lie just a little above the paragonite+quartz stability curve at low pressure and, because of the relative slopes, just below it when kyanite is a product. As shown on Fig. 4, this explains the phase relations mentioned above.

Evans (1965) found that paragonite alone breaks down at 590°C at 2.5 kbar (P_{H_2O}) and also showed the breakdown of muscovite is lowered 80° in the presence of quartz. Hence Reaction (12) in Figure 4 is placed at 510°C at 2 kbar. If this is even approximately correct, the preceding arguments suggest a similar location for Reaction 1.

As the assemblage sillimanite+paragonite is not found, it can be inferred that Reactions (11) and (12) lie at temperatures below the aluminosilicate triple point in contrast to Figure 4. However, the data given above indicates that it does not lie far below. Possibly only at high p_{H_2O} does it approach the sillimanite field.

GENERAL CONSIDERATIONS

Two basic controls have been suggested, one of which explains the general scarcity of paragonite (and pyrophyllite) and the second of which clarifies several of the restricted mineral associations of paragonite.

The first is a compositional control based on the relative Al_2O_3 content. Most pelites are too poor in Al_2O_3 to have paragonite until reactions such as (1) bring in kyanite and thereby release Al_2O_3 to the KNaAl system. The general incompatibility of paragonite+biotite has been emphasized (except in the assemblage kyanite+biotite+paragonite). Inasmuch as the vast majority of pelitic schists contain biotite, it is little wonder that paragonite is uncommon.

The second control (on restricted assemblages) involves a crossing of the *pT* curves for the upper stability of paragonite and the reaction first bringing in the pair aluminosilicate+biotite. The absence of andalusite+paragonite+biotite is thus explained. Also it suggests that in rocks of common bulk composition (below the chlorite-garnet join) those with kyanite+biotite+staurolite are very likely hosts to paragonite.

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[1] In the region between the two *pT* curves but below their intersection, Reaction (1) will actually result in the formation of a more sodic muscovite as well as the production of paragonite. Na would presumably be obtained from plagioclase, the pure Ab end member being theoretically unstable in the assemblage kyanite+muscovite+paragonite.

The most likely medium grade rocks to contain paragonite are those with high Al_2O_3 such as kyanite-quartzites, chloritoid+chlorite schists, or any rock with highly aluminous minerals and no biotite. In rocks of common bulk composition, which will develop biotite in low metamorphic grades, paragonite will usually be absent—even in an assemblage like staurolite+chlorite+biotite, because the bulk composition will still plot below the garnet-chlorite join. Only after reactions such as (1) bring in kyanite will paragonite be compositionally possible and then occur with biotite.

From Figure 4 the assemblage kyanite+staurolite+biotite+paragonite must occur over a very narrow temperature range. Rocks with kyanite+staurolite+biotite should always be investigated for paragonite. However this assemblage clearly extends above the stability of paragonite as Chinner (1965) and Phinney (1963) describe it without paragonite.

The foregoing remarks clearly show that it would be desirable to have the paragonite+quartz curve accurately determined in the laboratory—especially since complex reactions (1) seem to lie very close to it in terms of p and T.

ACKNOWLEDGMENTS

The writer is grateful to Professors A. L. Albee, C. Durrell, B. W. Evans, J. M. Moore, Jr., and E. M. Moores for their comments and criticisms on various drafts of this paper. This work was partially supported by NSF Grant GA-406. The writer is also grateful to Mrs. Ruth Darden for drafting Figures 1–4.

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Manuscript received, April 24, 1967; accepted for publication, January 3, 1968.