Alteration of andalusite to sheet silicates in a pegmatite

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ABSTRACT

Partially sericitized andalusite from pegmatites at Szabo Bluff, Scott Glacier, Antarctica, was investigated using transmission and analytical electron microscopy (TEM and AEM). Although muscovite and corundum occur as the major alteration products of andalusite, TEM and AEM investigations reveal that Al-rich chlorite exists as thin lamellae, 0.1- to 0.3-μm thick, within andalusite crystals. AEM analysis of the chlorite results in a formula of $Al_{4.32}Fe_{0.07}(Si_{2.83}Al_{1.17})O_{10}(OH)_8$, indicating that it is the di,dioctahedral chlorite donbassite. Similar Al/Si ratios of donbassite ($\simeq 1.94$) and andalusite ($\simeq 2.0$) and the textural relationship suggest that andalusite alteration to donbassite occurred through a hydration reaction, where an Al/Si ratio of $\simeq 2.0$ was nearly preserved. Andalusite was altered directly to either muscovite + corundum or to donbassite, depending on the availability of K^+ in the silica-deficient fluids. Donbassite apparently becomes unstable where K^+ is present, and it will eventually react to muscovite + corundum. The (001) planes of both sheet silicates are approximately parallel to {110} of andalusite, indicating that a cleavage-controlled alteration mechanism was dominant. Structural features of donbassite, such as severe stacking disorder and the common interlayering of other sheet silicates, suggest that donbassite may have formed metastably. However, the possibility of donbassite being a stable phase in the system Al_2O_3-SiO_2-H_2O cannot be excluded.

INTRODUCTION

Although andalusite typically occurs in contact-metamorphosed pelitic rocks, it has been reported from pegmatites (Webb, 1943; Grosemans, 1948; Rose, 1957; Voloshin and Davidenko, 1972; Burt and Stump, 1984). Such occurrences are restricted to relatively primitive and poorly fractionated pegmatites, and it is rare in spodumene-bearing or other complex pegmatite types (Černý and Hawthorne, 1982). The rarity of andalusite in igneous rocks is attributed to the extremely narrow pressure-temperature range over which andalusite, rather than muscovite or sillimanite, can grow from a water-saturated peraluminous granitic melt (Clarke et al., 1976; Burt and Stump, 1984).

Where andalusite occurs in pegmatites, it is invariably subject to late micaceous replacement by residual pegmatite fluids that produce a diverse array of alteration types (Černý and Hawthorne, 1982). Al-rich minerals in pegmatites are particularly easily altered to muscovite or more complex assemblages containing muscovite, and K^+ and H^+ metasomatism commonly results in muscovite replacing andalusite (Černý and Burt, 1984).

Muscovitization of andalusite is also common in metamorphic environments. Kwak (1971) noted that the susceptibility to muscovitization of Al_2SiO_5 polymorphs increases in the sequence of kyanite, andalusite, and sillimanite. He suggested that the different stability fields of Al_2SiO_5 polymorphs + solution under geologic conditions account for the selective muscovitization. On the other hand, Chinner (1973) suggested that selective alteration may be related to recrystallization history during metamorphic episodes.

When Al-rich minerals are altered by late fluids in pegmatites, they commonly react through various intermediate steps as the fluids evolve and as equilibrium is attained. For example, London and Burt (1982) showed that spodumene alters to eucryptite + albite, and eucryptite eventually alters to muscovite. Muscovite is usually the final alteration product. Although most andalusite from pegmatites is partially altered (Webb, 1943; Rose, 1957; Burt and Stump, 1984), detailed characterization of andalusite alteration has not been done. In this study, transmission electron microscopy and analytical electron microscopy have been used to examine mineralogical, chemical, and structural relations of minerals involved in the alteration of andalusite. Detailed investigation of andalusite alteration will provide additional insights into mineral reactions in the system Al_2O_3-SiO_2-$H_2O(\pm K_2O)$.
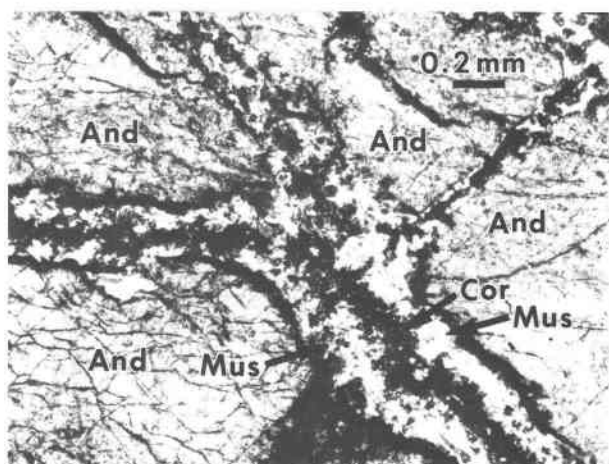
Fig. 1. Photomicrograph showing the replacement of anda-
lusite by muscovite. Corundum occurs within muscovite aggre-
gates. (Cross-polarized light.)

Fig. 3. TEM image of residual andalusite (A) lamellae within
muscovite (M).

EXPERIMENTAL METHODS

Thin sections were prepared using Crystalbond, a material that
can be melted by gentle heating and that is soluble in acetone.
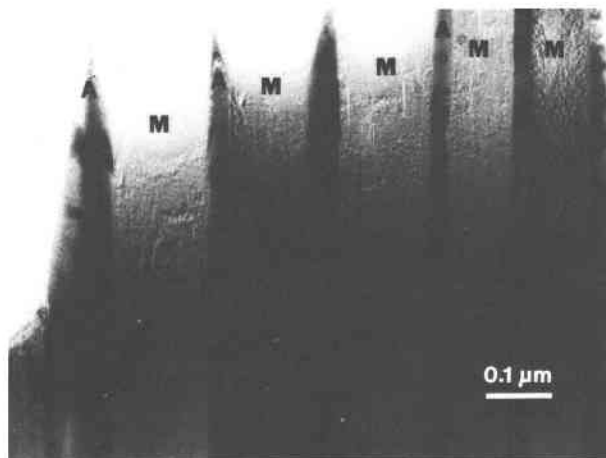After optical observation, selected areas were mounted with 3-mm
washers to make ion-thinned samples for transmission-electron-
microscope study. The washer-mounted specimens were de-
tached from the slides by heating, and they were ion-thinned
and carbon-coated. Most TEM imaging was done using a JEOL
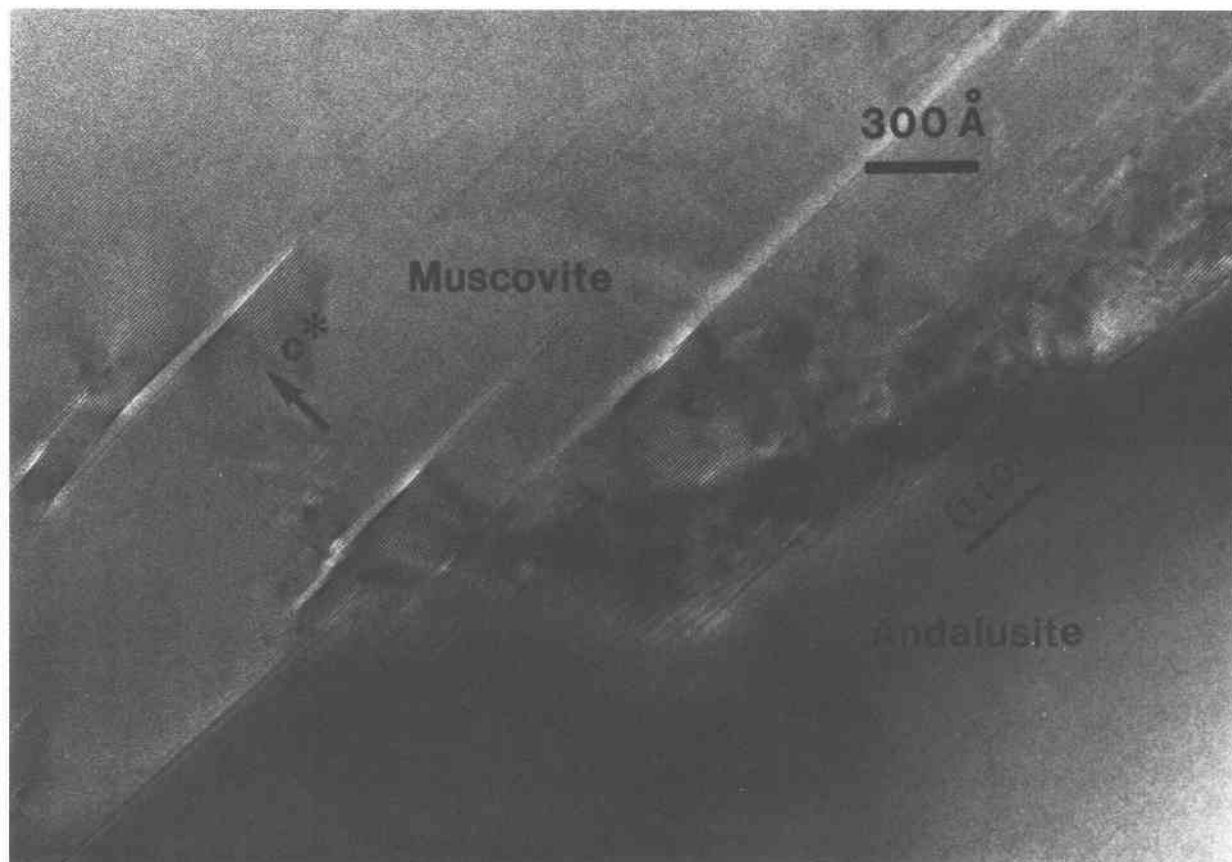200CX 200-kV electron microscope with top-entry stage having

Fig. 2. TEM image of the interface between andalusite and muscovite. Although interfaces are generally parallel to the muscovite-
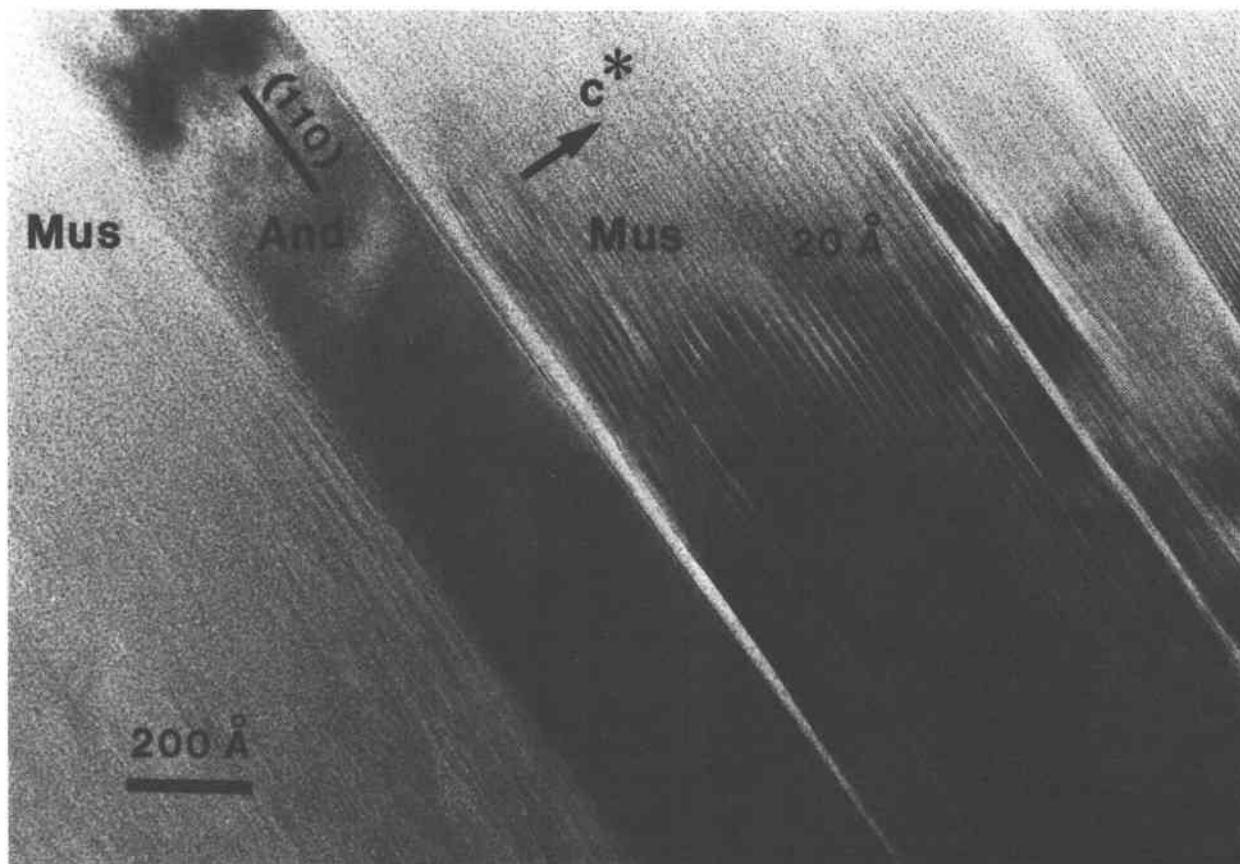layer direction, inclined interfaces also occur.

Fig. 4. TEM image of interface between andalusite (And) lamellae and muscovite (Mus). The (110) plane of andalusite is parallel to the muscovite-layer directions. Muscovite shows 20-Å lattice fringes because of *2M* polytypism.

tilting angles of $\pm 12°$, spherical aberration coefficient (C_s) of 1.2 mm, and structure resolution of 2.6 Å. AEM was performed using a Philips 400T 120-kV electron microscope with side-entry stage. It is equipped with a Gatan 607 electron energy-loss spectrometer and a Tracor Northern TN-2000 multichannel analyzer for electron energy-loss spectroscopy (EELS) and energy-dispersive X-ray analysis.

SPECIMENS AND GEOLOGIC SETTINGS

Andalusite was reported by Burt and Stump (1984) from the pegmatites at Szabo Bluff, Scott Glacier, Antarctica (lat 86°29′S., long 144°45′W.), that were discovered by Stump et al. (1981). Szabo Bluff appears to consist entirely of coarse-grained, K-feldspar–rich biotite granite. Three major and several minor podlike subhorizontal pegmatite bodies occur within the granite (Burt and Stump, 1984). The pegmatites have two easily recognized zones. Border zones are marked by coarse, dendritic to bladed biotite growing inward from the walls in a fine-grained to graphic feldspar-quartz matrix. Interior zones mainly consist of blocky K-feldspar and milky quartz with platy albite, minor tourmaline, and muscovite. Ilmenite and other oxides occur locally. Andalusite crystals are irregularly segregated as columnar, radiating, reddish-brown crystal aggregates in the interior zones of the two larger pegmatites. Some individual andalusite crystals are as long as 0.5 m.

PETROGRAPHIC MICROSCOPE OBSERVATIONS

The andalusite was described by Burt and Stump (1984) based on petrographic microscope observations. Andalusite is replaced by fine-grained muscovite along irregular veinlets (Fig. 1). The initial alteration apparently occurs along {110} cleavage directions of the host andalusite. Occurrence of muscovite by similar cleavage-controlled alteration of andalusite was also reported from pegmatites in Yosemite National Park, California (Rose, 1957). In addition, coarse-grained muscovite flakes are present as fracture-filling material.

Dark-blue corundum is commonly disseminated in the muscovite aggregates replacing andalusite (Fig. 1). Corundum does not occur inside or in contact with andalusite crystals. Diaspore was not observed in the altered andalusite, although Rose (1957) reported that rare diaspore occurs at the rims of corundum crystals.

TEM OBSERVATIONS

Andalusite alteration to muscovite

Muscovite, the major alteration product of andalusite, occurs mostly as subparallel aggregates. Although most
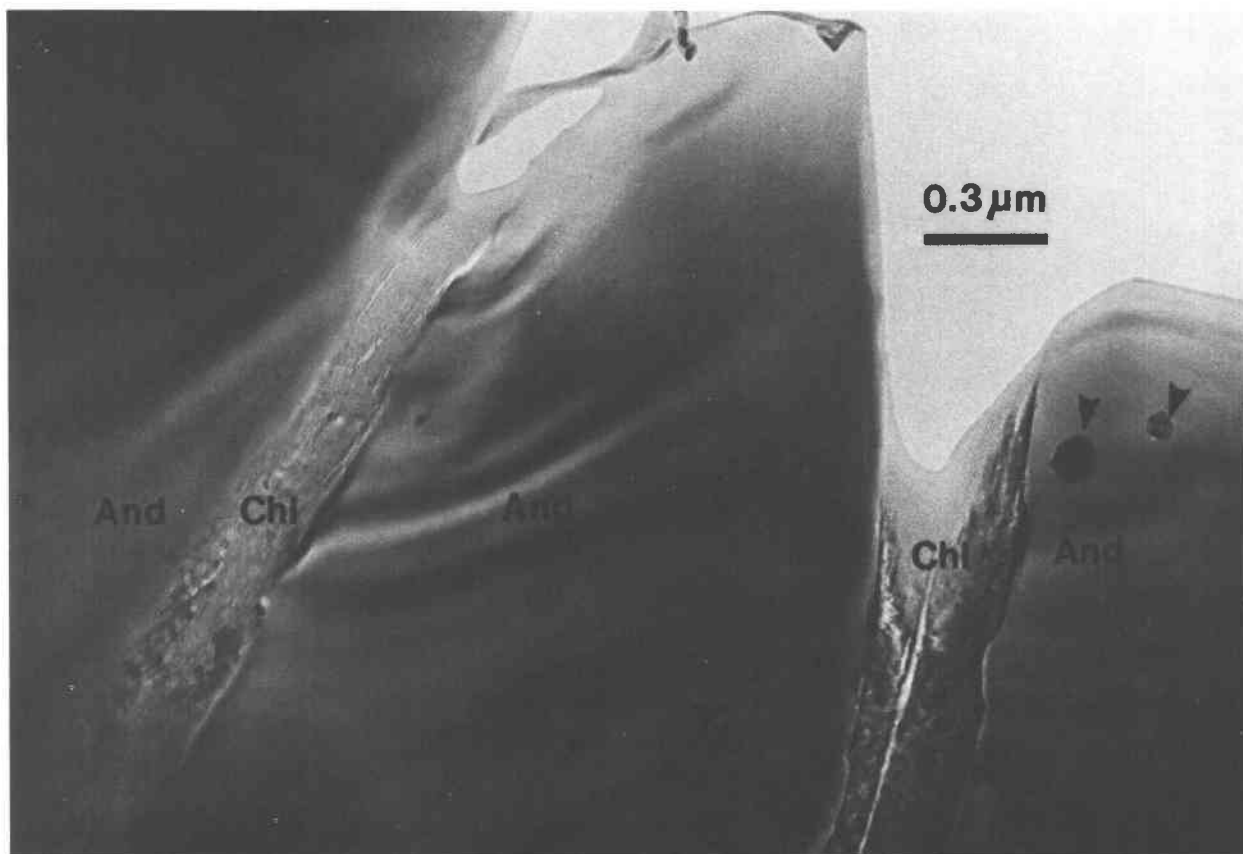
Fig. 5. TEM image showing the replacement of andalusite (And) by chlorite (Chl) lamellae. Carbon particles (indicated by arrows) were deposited during carbon-coating.

interfaces between muscovite and andalusite are parallel to the muscovite (001) layer direction, parts of interfaces are inclined to (001) of muscovite (Fig. 2). The exact boundaries become difficult to define when muscovite layers terminate at the inclined interfaces. Lenticular fissures of approximately 50-Å width, caused by electron-beam irradiation (Ahn et al., 1985, 1986), are common in the muscovite lattice-fringe images (Fig. 2). However, such beam damage is not as pervasive as in Na-rich micas (Ahn et al., 1986).

Unaltered andalusite remains within muscovite aggregates as narrow lamellae <0.1 μm wide. Thin edges show jagged shapes, indicating that andalusite is more resistant to ion-milling than muscovite (Fig. 3). (110) of andalusite approximately parallels (001) of the surrounding muscovite (Fig. 4), indicating that muscovite replaces andalusite mainly along the {110} cleavage.

All corundum occurs as thick, blocky crystals; it is apparently resistant to ion-milling compared to sheet silicates and andalusite. Fine-grained corundum crystals were not observed within muscovite aggregates; corundum may occur only as 0.02- to 0.03-mm anhedral crystals, as observed by optical microscopy (Fig. 1).

Andalusite alteration to donbassite

TEM investigation reveals that most andalusite crystals are partly replaced by chlorite, which occurs as 0.1- to 0.3-μm-thick lamellae (Fig. 5). AEM analyses of such chlorite indicate an average formula of $Al_{4.32}Fe_{0.07}$-$(Si_{2.83}Al_{1.17})O_{10}(OH)_8$ (see Table 1, Ahn and Buseck, 1988), showing that it is almost free of divalent cations such as Fe and Mg and that it is a near-end-member di,dioctahedral aluminous chlorite, donbassite.[1] Substitution of Al for Si in the tetrahedral sites results in excess Al in the octahedral sites. Electron-microprobe analysis of unaltered andalusite indicates an average formula of $Al_{1.99}Fe_{0.03}Si_{0.99}O_5$ (see Table 1, Ahn and Buseck, 1988), showing that similar amounts of Fe are present in andalusite and chlorite. The minor Fe in the donbassite apparently was derived from precursor andalusite.

TEM images indicate that the boundaries between an-

[1] The mineral name donbassite was recommended for chlorite with two dioctahedral sheets by the AIPEA and CMS Nomenclature Committees and IMA Commission on New Minerals and Mineral Names (Bailey et al., 1979; Bailey, 1987, pers. comm.).
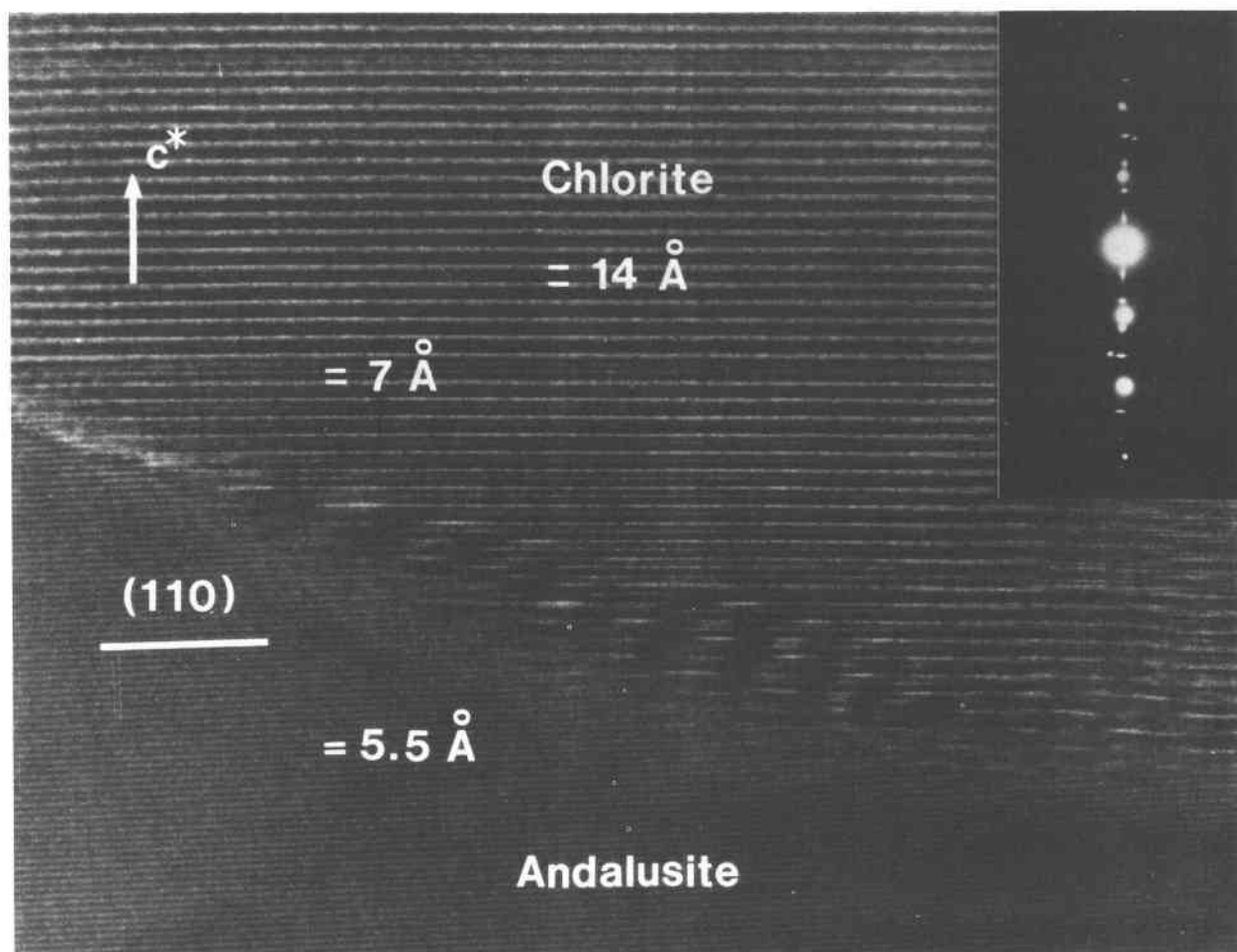
Fig. 6. TEM image of interface between andalusite and chlorite (donbassite). The (110) plane of andalusite and the (001) plane of chlorite are approximately parallel. Isolated 7-Å layers occur as intercalation defects.

dalusite and donbassite are irregular (Fig. 6), although (001) of chlorite and (110) of andalusite are approximately parallel. Lattice fringes of chlorite at the interfaces exhibit different contrast and slight bending. This material may represent incipient donbassite that is poorly crystallized at the reaction boundaries. Alternatively, such contrast change may result from the presence of strain or layer distortion produced by structural misfit or incoherency between the donbassite and andalusite structures. Layers having 7-Å spacings are commonly interlayered with donbassite (Fig. 6).

Microstructures of sheet silicates

Selected-area electron diffraction (SAED) indicates different degrees of stacking disorder in both sheet silicates. SAED patterns of donbassite exhibit heavy streaking in the reciprocal rows with $k \neq 3n$ and sharp reflections with $k = 3n$ (Fig. 7a). These features indicate that stacking disorder is mainly caused by a shift of $\pm b/3$ or by random layer rotation by $n(120)°$ between alternating pyrophyl-lite-like and gibbsite-like layers. Such stacking disorder is common in chlorite and micas (Brown and Bailey, 1962; Brindley, 1980). On the other hand, all muscovite in this study shows two-layer polytypism, and [110] SAED patterns exhibit relatively sharp reflections without distinct streaking in the reciprocal rows with $k \neq 3n$ and $k = 3n$ (Fig. 7b), indicating that stacking disorder in muscovite is minor or absent.

Donbassite is commonly interlayered with other sheet silicates (Fig. 8); 7-Å layers are most abundant, mainly as single layers. AEM examination of areas rich in such 7-Å layers does not indicate high concentrations of Fe or Mg, suggesting that the 7-Å layers may be kaolinite. Interlayering of 7-Å layers with chlorite is only reported from trioctahedral chlorites from diagenetic or very low grade metamorphic environments (Lee et al., 1984; Ahn and Peacor, 1985). Such 7-Å layers interlayered with trioctahedral chlorites were characterized as berthierine, which is chemically similar to Fe-rich trioctahedral chlorite (Ahn and Peacor, 1985). Besides 7-Å layers, 9.3-Å
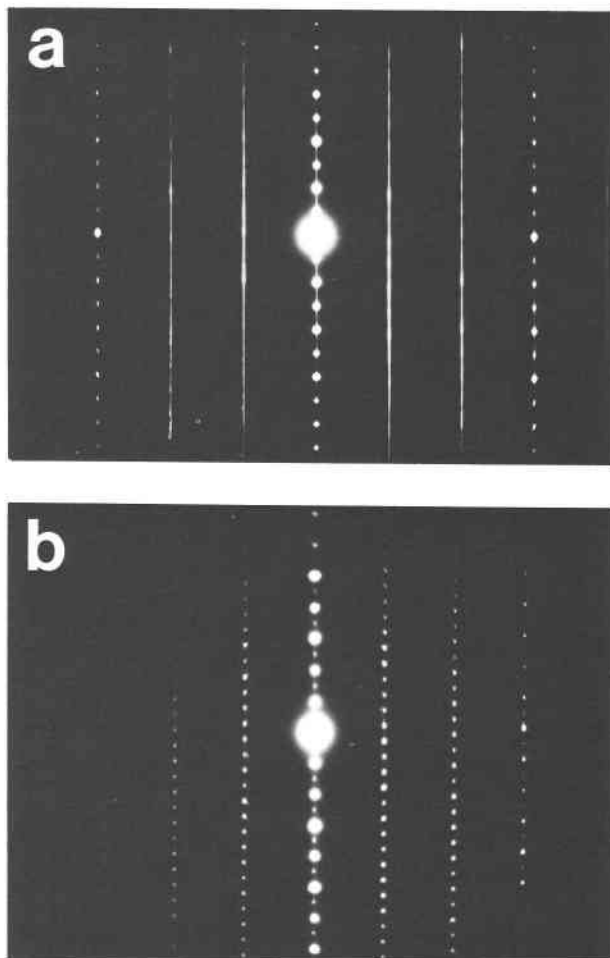
Fig. 7. [110] SAED patterns of (a) donbassite and (b) muscovite.

layers occur in places. The 9.3-Å interlayer spacings suggest that they are pyrophyllite-like layers rather than muscovite. The intercalation of 9.3-Å layers may arise from the absence of gibbsite-like layers or the presence of extra pyrophyllite-like layers in the donbassite structure, similar to isolated talc-like layers found in the trioctahedral chlorite that occurs as an alteration product of a biotite (Veblen, 1983). However, extra gibbsite-like layers were not seen in the donbassite crystals.

In contrast to donbassite, muscovite does not show interlayering of other sheet silicates. The possible presence of pyrophyllite instead of muscovite was checked by AEM analysis, and no 2:1 layer silicates other than muscovite were detected.

DISCUSSION

Alteration of andalusite

The possible existence of chlorite having two dioctahedral layers was predicted by Brindley and Gillery (1956) and Grim (1968). Relatively pure aluminous chlorite has only been reported from the hydrothermal ore veins of

the Donetz Basin, USSR (see the review by Rozinova and Dubik, 1983) and was named donbassite (Lazarenko, 1940). Less well-crystallized layer silicates comparable to di,dioctahedral chlorite were reported as components of some mixed-layer clays or as a mixture with other minerals in sedimentary rocks (Eggleton and Bailey, 1967; Müller, 1967; Bayliss and James, 1981).

The Al/Si ratio of donbassite (1.94) is approximately similar to that of andalusite (2.01), indicating that alteration occurred through a hydration reaction in which the Al/Si ratio was nearly preserved. Andalusite to donbassite alteration may have been facilitated by the similar Al/Si ratio of both minerals under the conditions of low silica activity at the incipient alteration stage. The susceptibility of alteration of aluminous minerals to other sheet silicates having the same Al/Si ratio has been shown in eucryptite to muscovite alteration, where an Al/Si ratio of $\simeq 1.0$ is retained (London and Burt, 1982). Another example of alteration of andalusite to sheet silicates of Al/Si ratio of $\simeq 2.0$ occurs in margarite pseudomorphs of andalusite in metamorphic rocks (Velde, 1971; Guidotti and Cheney, 1976; Guidotti et al., 1979). In the metapelites in the Georgetown area, California, muscovite and paragonite occur as matrix surrounding the margarite pseudomorphs after andalusite (Guidotti et al., 1979). Coexistence of muscovite and paragonite as a matrix surrounding margarite pseudomorphs indicates that replacement of andalusite by margarite was favored by their similar Al/Si ratios ($\simeq 2.0$) despite the presence of K^+ and Na^+ in the fluids, so that only CaO and H_2O were added (Guidotti et al., 1979).

Muscovite and corundum constitute the major alteration products of andalusite. Andalusite to muscovite + corundum alteration has been proposed by Rose (1957) and Burt and Stump (1984) following the reaction 6 andalusite + $2K^+$ + $3H_2O$ = 2 muscovite + 3 corundum + $2H^+$. Corundum occurs exclusively within muscovite aggregates, suggesting that corundum is also an alteration product rather than a primary mineral. Rose (1957) indicated that highly aluminous phases such as corundum or diaspore form as by-products of the andalusite to muscovite reaction in locally silica-deficient environments. Silica from the pegmatite is apparently unable to migrate to the interiors of the andalusite crystals to participate in the alteration process. The silica deficiency mainly results from the different Al/Si ratio of andalusite ($\simeq 2.0$) and muscovite ($\simeq 1.0$). Such reaction relationships are in contrast to the muscovitization of andalusite and other aluminum silicates in metamorphic rocks, where corundum is absent because of the excess of silica in that environment. Kwak (1971) suggested a muscovite-formation reaction: 3 andalusite + $3SiO_2$ + $2K^+$ + $3H_2O$ = 2 muscovite + $2H^+$. K^+ and H^+ metasomatism is involved in this reaction as well as in the andalusite to muscovite reaction in silica-undersaturated environments.

The dominant occurrence of muscovite indicates that andalusite alteration to muscovite + corundum rather than to donbassite was caused by the presence of K^+ in
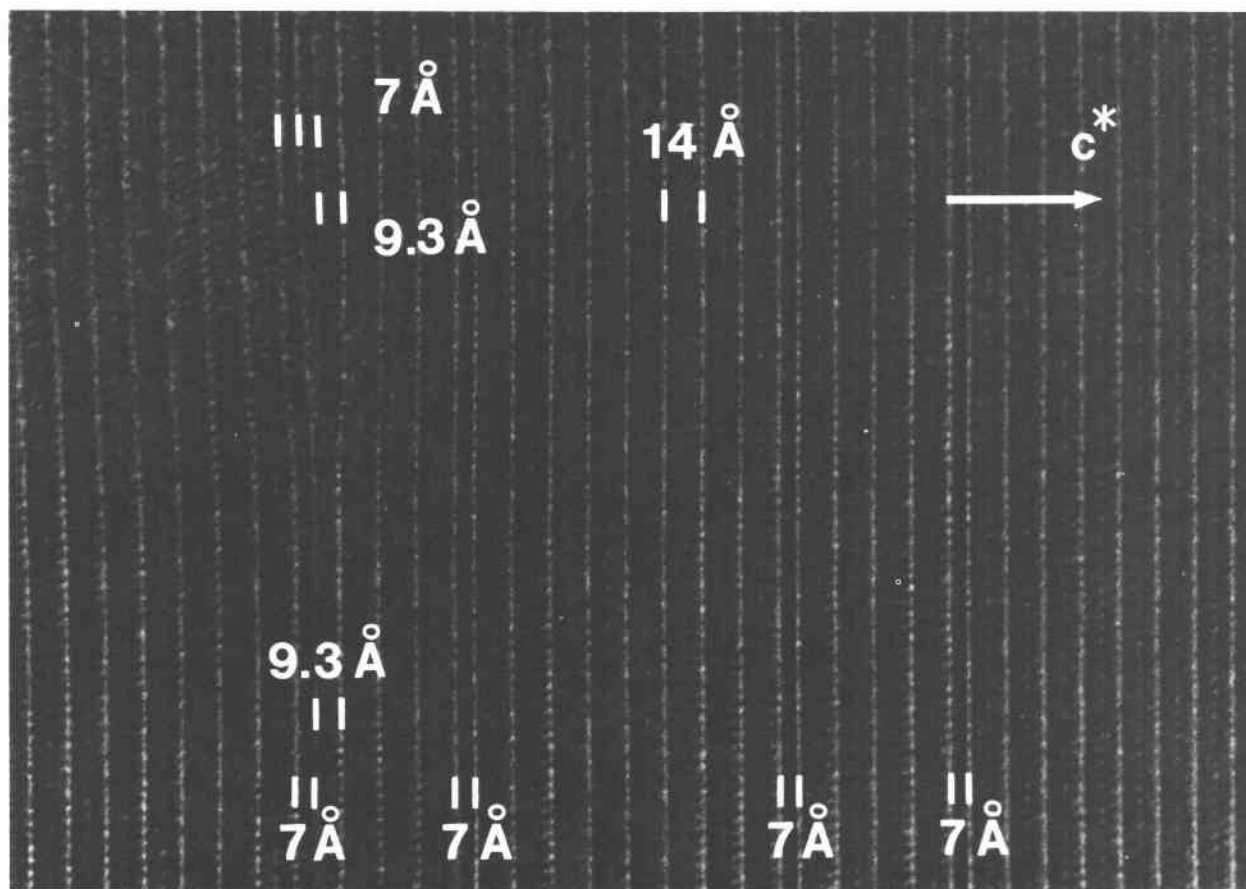
Fig. 8. TEM image of intercalation features in donbassite. 7-Å layers are more common than 9.3-Å layers. An edge dislocation is present as a result of the presence of an extra 7-Å layer.

the fluids. It is also possible that end-member donbassite is itself metastable with respect to assemblages of other aluminous phases such as diaspore, pyrophyllite, or kaolinite. Such phase relationships may be supported by the disequilibrium mineral textures showing common interlayering of 7-Å layers (kaolinite-like layers) and 10-Å layers (pyrophyllite-like layers). Similar disequilibrium textures were seen in the hydration reaction of chain silicates to serpentine and talc (Veblen and Buseck, 1979).

Boundaries between donbassite and muscovite were not observed in this study, although they might be expected at the boundaries between muscovite and partially chloritized andalusite crystals. In addition, donbassite was not observed within muscovite aggregates. Donbassite apparently becomes unstable where K^+ is available, and donbassite reacted to muscovite + corundum in the same way as andalusite reacted to muscovite + corundum. Such alteration trends are generalized in Figure 9. The bulk Al/Si ratio of $\simeq 2$ was apparently preserved in all of these alteration reactions.

The (001) planes of both donbassite and muscovite are generally parallel to {110} of host andalusite. Examination of the [001] projection of the andalusite structure

indicates that andalusite is not likely to have coherent boundaries with chlorite or muscovite. The unusual contrast of lattice fringes at the interface (Fig. 6), where (110) of andalusite and (001) of donbassite are parallel, may indicate the presence of strain because of the structural misfit. The petrographic microscope observations of {110} cleavage-controlled replacement are consistent with these TEM observations and suggest that preferential replacement of sheet silicates was favored by the dominant {110} andalusite cleavage. Similar cleavage-controlled alteration also occurs in spodumene (London and Burt, 1982).

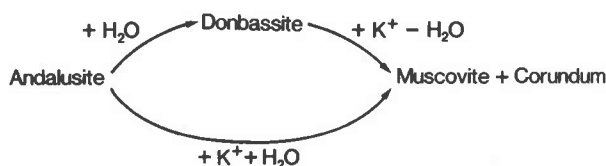Aluminous minerals having an Al/Si ratio of 2, such as

Fig. 9. Generalized alteration sequence for andalusite. Andalusite reacts to muscovite + corundum directly or through donbassite.
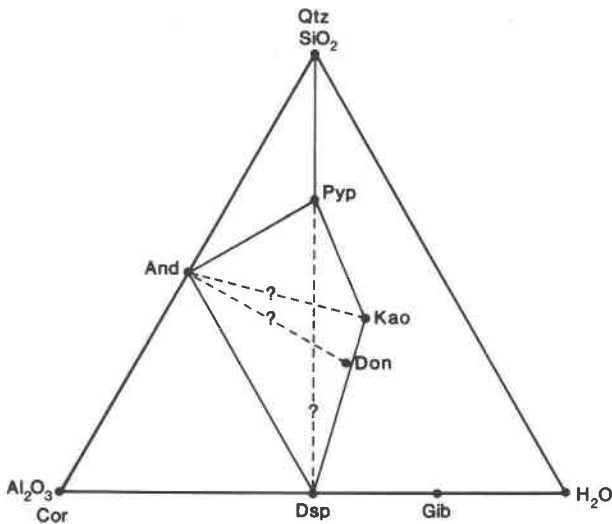
Fig. 10. Composition diagram in the system Al_2O_3-SiO_2-H_2O. Abbreviations: Don = donbassite, And = andalusite, Cor = corundum, Dsp = diaspore, Gib = gibbsite, Kao = kaolinite, Pyp = pyrophyllite, Qtz = quartz.

sillimanite, kyanite, or topaz, could also alter directly to donbassite by hydration, given a sufficiently alkali- and silica-deficient environment. Otherwise they would alter to muscovite (+ corundum, if silica is deficient), biotite, margarite, or ephesite depending on the fluid chemistry. Although Wenk (1980) reported disordered chlorite in the vicinity of kyanite-staurolite interfaces, it was a trioctahedral chlorite. The apparent scarcity of donbassite may in part be caused by the difficulty of identifying it since it has the same Al/Si ratio as its anhydrous precursor minerals (Ahn and Buseck, 1988).

Phase equilibria

Whether or not the donbassite replacing andalusite formed stably or metastably remains an open question. No previous discussion of mineral stabilities in the system Al_2O_3-SiO_2-H_2O (e.g., Zen, 1961; Day, 1976; Burt, 1978; Perkins et al., 1979; Stout, 1985) has considered its possible stability or even existence, nor has it been reported in experimental run products (e.g., Roy and Osborn, 1954; Haas and Holdaway, 1973; Hemley et al., 1980).

The composition of donbassite lies between that of andalusite and H_2O, as expected from the fact that it is a hydration product of andalusite (Fig. 10). Its composition also lies nearly between that of diaspore (AlO(OH)) and kaolinite. There is a possible H_2O deficiency relative to the linear combination of diaspore and pyrophyllite that results from the coupled substitution $Al_4(Si_3\square)_{-1}$. Donbassite compositions must lie along a line between kaolinite and a theoretical end member $(Al_{5.33}\square_{0.67})$-$Al_4O_{10}(OH)_8$ having an Al_2O_3:H_2O ratio of 7:6 instead of the 1:1 of diaspore, thereby explaining why donbassite lies slightly off the kaolinite and diaspore tie line (Fig.

10). Adding the additional constraint that the Al:Si ratio is 2:1 yields a donbassite composition of $(Al_{4.4}\square_{1.6})$-$(Si_{2.8}Al_{1.2})O_{10}(OH)_8$, which is extremely close to the composition that we measured.

Most of the above-cited studies have assumed that diaspore + pyrophyllite is a stable assemblage, which implies that the stable reaction of andalusite to donbassite is unlikely. Nevertheless, andalusite + kaolinite assemblages were reported by Zen (1961) and Sykes and Moody (1978); furthermore, Day (1976), Perkins et al. (1979), and Stout (1985) consider it possible that andalusite + kaolinite form a stable assemblage at relativley low pressures relative to diaspore + pyrophyllite. If the diaspore + pyrophyllite tie line can be broken, then the hydration of andalusite to donbassite could represent a stable reaction, assuming the donbassite itself is stable. In any case, the kaolinite + pyrophyllite tie line excludes donbassite from silica-saturated assemblages.

Experimental hydrothermal studies and precise thermodynamic characterization of donbassite are necessary to resolve the question of its stability relative to the other phases in the system and to determine the influence of minor Fe on its stability. Additional natural occurrences should also be sought. Judging from the phase diagram, the best places to look would be in diaspore- and kaolinite-bearing rocks formed at low pressure and temperature, or where aluminosilicate minerals are replaced in silica-undersaturated rocks.

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