

High-resolution Transmission Electron Microscopy of Tremolite-to-Talc Reaction at the Dongyang Talc Deposit

동양 활석광상에서의 투각섬석-활석 반응에 관한 고분해능 투과전자현미경학적 연구

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ABSTRACT: Tremolite crystals from the Dongyang talc deposit were studied using high-resolution transmission electron microscopy (HRTEM) to characterize the tremolite-to-talc reaction. [001] HRTEM images of tremolite show intergrowths of wide-chain pyriboles and talc; talc is the primary alteration product of tremolite, and triple-chain structures occur sparsely. The boundaries between tremolite and talc are commonly well defined by (010) and (100) interfaces. (001) talc layers are parallel to (100) of tremolite, and the interfaces between tremolite and talc appear to be coherent in HRTEM images, indicating that most talc layers formed directly from tremolite by a hydration reaction. However, some talc formed along (110) of tremolite, and talc layers are not extended from (010) of tremolite, suggesting that part of talc in the deposit was produced through a dissolution-precipitation mechanism. Carbonate minerals are also associated with tremolite and talc. Common replacement of dolomite by calcite indicates that the tremolite-to-talc reaction results in remnant Ca, which was eventually consumed to replace dolomite to form calcite. Some Mg produced by dolomite during reaction to calcite was apparently utilized to form talc, because talc formation from tremolite requires extra Mg. Although talc could be formed directly from dolomite, extensive alteration of tremolite to talc suggests that part of talc of the deposit was produced from tremolite that was formed by dolomite reaction during an early stage metamorphism.

Keywords: tremolite, talc, pyriboles, Dongyang talc deposit, high-resolution transmission electron microscopy (HRTEM)

요약: 동양 활석광상에서 산출하는 투각섬석에서의 투각섬석-활석 반응에 관한 연구를 위하여 고분해능 투과전자현미경을 이용하여 관찰하였다. 투각섬석의 [001] 방향에서 관찰한 결과 다쇄구조 파이리볼(pyribole)들과 활석이 투각섬석 결정내에 협재되어 있음이 발견되었다. 활석이 투각섬석의 주된 변질물질로 관찰되지만, 파이리볼은 매우 소량 나타난다. 투각섬석과 활석은 주로 투각섬석의 (010)와 (100) 경계면을 따라서 관찰된다. 활석의 (001)은 투각섬석의 (100)와 평행하며 활석과 투각섬석의 경계면에서 결정구조의 연속성이 이루어지는 특징은 활석이 투각섬석으로부터 직접 수화작용에 의하여 생성되었음을 가리킨다. 그러나, 투각섬석의 (110) 면을 따라서 산출하는 일부 활석은 투각섬석과의 구조적인 연속성이 결여되어 있으므로, 이러한 특징은 일부 활석은 용해와 침전 작용

에 의해서 생성되었음을 지시한다. 탄산염광물들도 투각섬석 및 활석과 밀접하게 관련되어 산출한다. 투각섬석이 활석으로 반응하면서 방출되는 Ca는 백운석 결정이 방해석으로 교대되면서 소모되며, 방해석이 백운석을 교대하면서 방출되는 Mg은 투각섬석으로 부터 활석이 생성되면서 사용된 것으로 보인다. 이 연구에서 관찰되는 투각섬석의 활석화작용은 활석들이 백운석으로 부터 직접 생성될 수도 있지만 일부 활석은 백운석의 변성작용으로 생성된 투각섬석으로 부터 생성되었음을 지시한다.

주요어: 투각섬석, 활석, 파이리톨, 동양 활석광상, 고분해능 투과전자현미경

Introduction

Dongyang talc deposit, which is one of most important talc deposits in Korea in its quality and production, is emplaced in the Hyangsanri Dolomite near the Chungju area, northwestern part of the Okchon metamorphic belt. Kim *et al.* (1963) reported that Dongyang talc deposit formed as a hydrothermal replacement deposit by ascending residual fluids following the intrusion of amphibolite. Reedman *et al.* (1973) reported that the deposit formed by regional metamorphism and that the related hydrothermal fluids was associated with the intrusion of Jurassic granodioritic complex. Moon and Kim (1988) interpreted that the origin of hydrothermal fluids causing this deposit is not related to contact metamorphism or regional metamorphism. In addition, Park *et al.* (1995) concluded that the talc deposit formed as replacement deposit prior to the latest phase deformation that Okchon Metamorphic Belt has undergone.

Although many workers investigated the Dongyang talc deposit in detail, the timing of mineralization and the origin of the ore fluid are still in discussion (e.g., Kim *et al.*, 1963; Reedman *et al.*, 1973; Lee, 1987; Moon and Kim, 1988; Kim and Cho, 1993; Park *et al.*, 1995). High-resolution transmission electron microscopy (HRTEM) was utilized in this study to reveal tremolite-to-talc reaction, which is apparently one of important mineral reactions contributed to form the Dongyang talc deposit (Park *et al.*, 1995). HRTEM can provide structural details below the unit-cell level of most minerals, and therefore mineral reaction

mechanisms that occur only on a few angstrom scale can be imaged. Especially chain and sheet silicates were investigated extensively by many workers using HRTEM more than any other mineral groups (see the reviews by Buseck *et al.*, 1980; Veblen, 1981, 1992), partly because these minerals commonly show various non-periodic structural modifications and fine-scale intergrowths.

Microstructural variations and alteration characteristics of anthophyllite-talc were discussed by Veblen and Buseck (1979), and synthetic tremolite was studied by Maresch and Czank (1983, 1988) and Ahn *et al.* (1991). However, microstructural details of tremolite-to-talc alteration was not investigated in detail despite their importance in mineralogy and petrology. Isotope study of Dongyang talc deposit by Park *et al.* (1995) reported that part of talc ores formed from tremolite and that the tremolite-to-talc reaction is related to the reaction of associated carbonate minerals. We have studied the talc and tremolite crystals emplaced in the talc deposit to examine the microstructures of silicates related to the talc formation mechanisms. Our HRTEM study emphasized on the structural details found at the tremolite-talc interfaces, because talc formation mechanisms could be explained by structural changes at the reaction boundaries.

Experimental Methods

Talc-tremolite aggregate specimens investigated in this study were collected from the Dongyang talc deposit, which is embedded in

Hyangsanri Dolomite near Chungju area. The samples used in this study were collected from the rim area of the talc deposit, and they are identical to the specimen NH4 of Seoul National University collection. The general geological outline of the Dongyang talc deposit area is available in Lee (1987), Moon and Kim (1988), Kim and Cho (1993), and Park *et al.* (1995).

Samples were prepared as polished thin sections oriented approximately perpendicular to [001] of tremolite to observe tremolite crystals along two-cleavage direction. General characteristics of mineral texture were investigated using back-scattered electron (BSE) imaging method with electron probe microanalyzer (EPMA) as well as petrographic microscope. Selected areas for TEM observation were covered by 3-mm copper grids using epoxy glue. The grid-mounted specimens were detached from glass slides by gentle heating and were cleaned using acetone prior to ion-milling. Ion-milled specimens were investigated using a JEOL JEM-4000EX transmission electron microscope with a top-entry stage having tilting angles of $\pm 15^\circ$, spherical aberration coefficient (Cs) of 1.0 mm, and structure resolution of 1.7 Å (Smith *et al.*, 1986). A 40- μm objective aperture and a 150- μm condenser aperture were used for HRTEM imaging.

Imaging of the tremolite specimens was performed with the *c* axis (or [001]) parallel to the electron beam. [001] images of pyriboles were interpreted based on the image simulation results of Veblen and Buseck (1979) who showed that the positions of the "I-beam" units of pyriboles correspond to the dark areas for a wide range of underfocus conditions. The white spots in [001] images match with the A sites that have low or no electron density; the chain width and disposition of intergrown pyribole units were characterized based on the position and size of these spots. In addition, lattice-fringe images were obtained from sheet silicates, such as talc and chlorite, intergrown with tremolite, and were characterized by their interplanar spacings.

Results

The degree of alteration of tremolite to talc varies from crystal to crystal. Some tremolite occurs as euhedral crystals, and fine-grained talc replaces tremolite (Fig. 1a). Fine talc veinlets develop approximately along {110} cleavage direction, but part of tremolite is replaced by talc irregularly. Figure 1a shows that tremolite is replaced by talc along the cleavage directions and irregularly from rim area. Furthermore, fine-grained calcite occurs along with talc within tremolite crystals. These alteration features indicate that alteration of tremolite has progressed along {110} as well as irregularly from rim area and that tremolite reacted talc and calcite. Dolomite crystals were also found within talc (Fig. 1b). Dolomite crystals occur without distinct crystal shape, and the rim area of dolomite is commonly replaced by calcite.

Low-magnification TEM image also reveals that tremolite crystals are replaced by talc along cleavage direction (Fig. 2a), in consistent with the BSE image observation (Fig. 1a). The width of talc veinlets along cleavages is approximately 1 μm , and thin talc veinlets are further developed irregularly along fractures. Figure 2b shows interfaces between tremolite and talc at low magnification. The interfaces are sharp and well-defined, and they show stepwise characteristics, suggesting that the tremolite-to-talc alteration is controlled by the structures of both minerals.

HRTEM investigation revealed that tremolite crystals do not contain abundant wide-chain structures, but part of some crystals shows intergrown triple- and quintuple-chain structures (Fig. 3). Wide-chain slabs are extended along \mathbf{a}^* , and the chain width of the intergrown structures can be determined from the width of white area along the \mathbf{b} direction. Triple chains are most abundant among the intergrown chain silicates in tremolite crystals, and quintuple chains are rarely found (Fig. 3). Wide chains are intergrown irregularly as isolated slabs

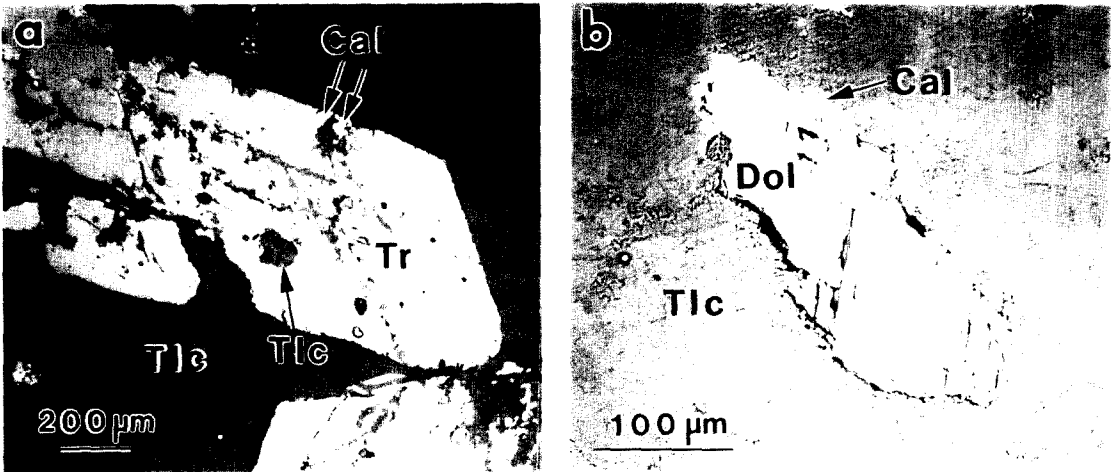


Fig. 1. BSE images tremolite and dolomite crystals within talc. (a) Euhedral tremolite (Tr) crystals are partially altered to talc (Tlc) with darker contrast and calcite (Cal) with brighter contrast. Talc replaces tremolite along {110} cleavage of tremolite. (b) Dolomite crystals found within talc. Note that calcite partially replaces dolomite (Dol) and that calcite shows brighter contrast than dolomite as a result of higher average atomic number.

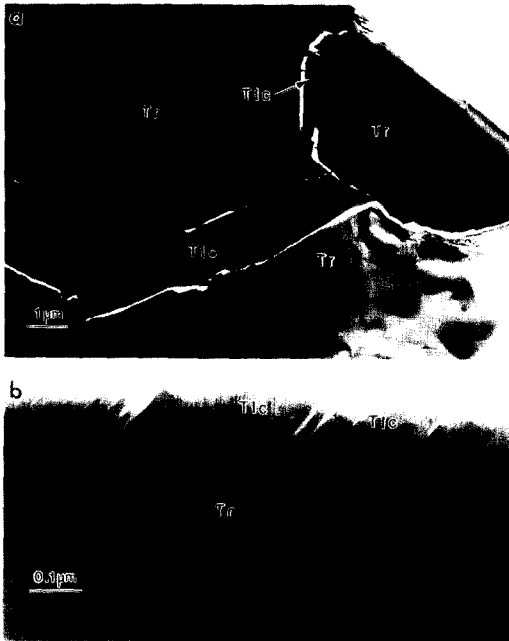


Fig. 2. Low-magnification TEM image showing alteration textures of tremolite (Tr). (a) Tremolite crystals are replaced by talc (Tlc) along cleavage direction and irregularly along fractures. (b) The interfaces between tremolite and talc show sharp stepwise characteristics.

along (010) within the tremolite crystals, resulting in "chain-width defects" (Veblen, 1981). The chain-width defects do not cause any structural distortion within the crystals and the intergrown wide chains maintain structural coherency with tremolite (Fig. 3).

Talc consists of subparallel packets of 9.3-Å layers (Fig. 4). Talc layers are not perfect straight layers, which is characteristic of micas, but they consist of somewhat imperfect and curvy layers. Low-angle grain boundaries are common, and packets of talc layers intersect subparallel layers at the boundaries. Each packet of layers has a slightly different orientation, and low-angle grain boundaries form where such packets coalesce. Talc grains are relatively free of other minerals other than chlorite in HRTEM observation. Figure 5 shows that thin packets of chlorite layers whose interplanar spacing is approximately 14 Å are intergrown within talc. Chlorite layers are oriented parallel to (001) of talc, and they are rare within talc.

HRTEM images show that the interfaces between tremolite and talc show various

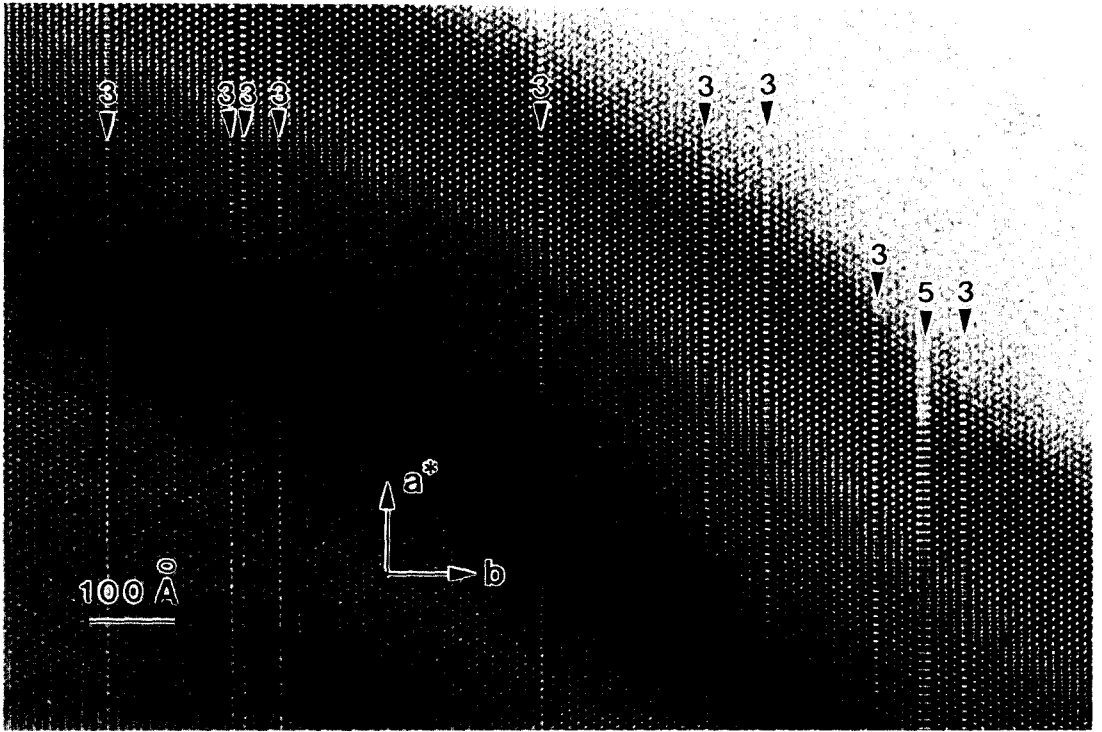


Fig. 3. [001] HRTEM image of tremolite that contains intergrown wide-chain pyriboles within otherwise regularly ordered double-chain structure. The integers marked on the image indicate the chain width of intergrown pyriboles.

characteristics. The boundaries between tremolite and talc are commonly well defined by (010) of tremolite, and (001) of talc is parallel to (100) of tremolite (Figs. 6a and 6b). In addition, it is also noted that (100) of tremolite could constitute well-defined boundary with talc (Fig. 6b). Development of both (010) and (100) interfaces results in the stepwise boundary that was presented in Fig. 2b. In some instances, the boundary between tremolite and talc is irregular and diffuse (Figs. 6c and 6d). The lattice fringes of talc partly overlap at the interfaces with the "I-beam" images of tremolite (Fig. 6d), suggesting that the boundary between these two minerals is not parallel to [001] of tremolite. However, (001) of talc is parallel to the *c* axis of tremolite in HRTEM images, indicating that the chain direction of tremolite is parallel to the talc layers. In general, (001)

of talc layers are not perfectly parallel to (100) of tremolite, and few talc layers are missing at the interfaces with tremolite. Such imperfection is apparently caused by slight dimensional difference between the "I-beam" units of tremolite and talc layer along the (010) interface.

Talc is mostly developed along the (010) and (100) boundaries of tremolite, and (001) of talc is parallel to (100) of tremolite. However, talc could develop along (110) of tremolite (Fig. 7). It is also noted that such interfaces are not well-defined, and stepwise ledges are present at the (110) boundary of tremolite. The talc layers are not continuous along (010) of tremolite, but (001) of talc layers is in general parallel to (110) of tremolite. Such texture is indicative of the talc formation mechanism that all talc did not formed topotactically from tremolite through solid-state transformation.



Fig. 4. HRTEM image of talc composed of subparallel packets of talc layers. Packets of talc layers intersect subparallel layers at the boundaries, resulting in low-angle grain boundaries.

Tremolite crystals contain holes, and HRTEM observations further revealed that part of the holes is commonly filled with "wormy-shaped" sheet silicates whose interplanar spacing is approximately 9.3 Å (Fig. 8). The interplanar spacing of such sheet silicate suggests that it is talc. The talc layers are partly extended from the tremolite "I-beam" units, and (001) of talc is in general parallel to (100) of tremolite. These characteristics indicate that talc replaces tremolite topotactically, and it is thus likely that such talc layers formed directly from tremolite during alteration. The holes apparently represent the alteration features produced by dissolution.

The size of the holes within tremolite could be variable, and wide-chain structures are associated with the talc in some area (Fig. 9). A quintuple chain terminates at one end and another quintuple chain occurs on the opposite side by taking a step in the *b* direction; the total number of chains are

identical on both sides of the termination, maintaining structural consistency with neighboring double-chain units of tremolite. Such a termination of wide-chain structure with the same chain numbers on both sides is referred to as "cooperative zipper termination" (Veblen and Buseck, 1980). Further details on theoretical relationships involved in the zipper termination are discussed in Veblen and Buseck (1980).

Discussion

Tremolite crystals embedded within talc show that fine-grained talc replaces tremolite (Fig. 1a), and low-magnification TEM images also reveal that tremolite crystals are replaced by talc along cleavage direction (Fig. 2a), suggesting that a cleavage-controlled alteration mechanism is associated with tremolite-to-talc reaction. However, thin talc veinlets were

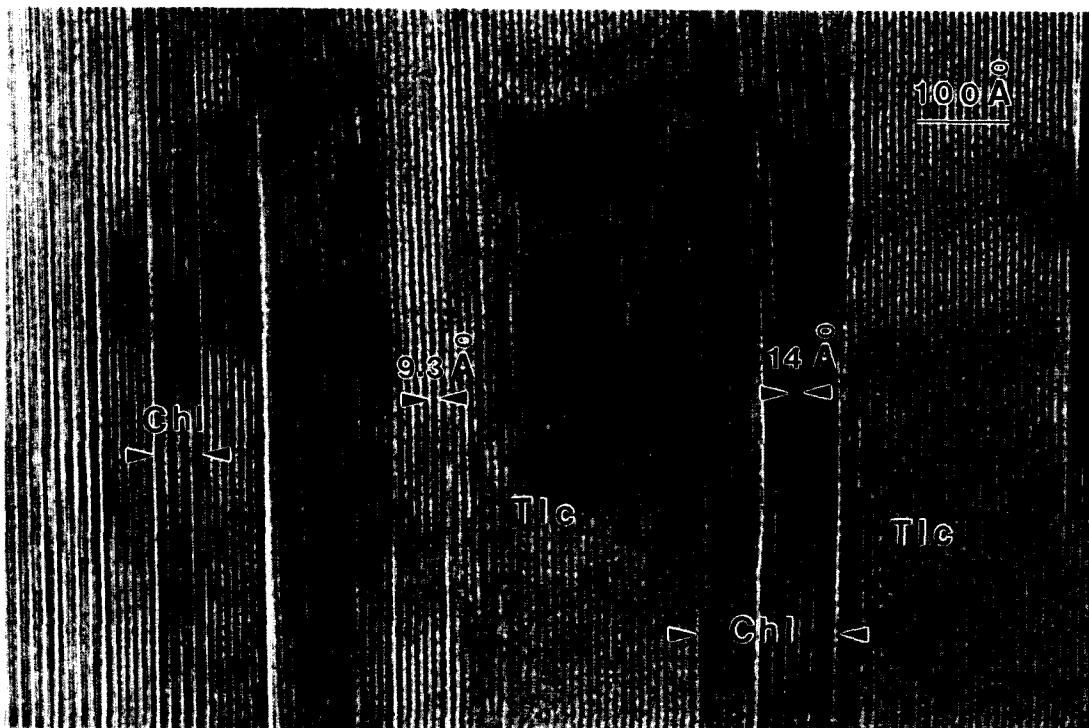


Fig. 5. Thin packets of chlorite (Chl) layers are locally intergrown within talc. Chlorite layers are oriented parallel to (001) of talc.

developed irregularly along fractures (Fig. 2a), and fracturing of tremolite may also played an important role in the talc formation. Moon and Kim (1988) suggested that the origin of hydrothermal fluids causing this deposit is not related to contact metamorphism or regional metamorphism and that deformation such as slight fracturing occurred prior to mineralization.

The common occurrence of the (010) and (100) interfaces between tremolite and talc as well as structural continuity of these two minerals indicate that talc formed from tremolite by hydration reaction of tremolite. The (001) layers of talc are parallel to (100) of tremolite, and the interfaces between tremolite and talc appear to be coherent in HRTEM images. It is thus likely that talc layers formed at the expense of tremolite by a hydration reaction. The $d(001)$ of talc, $\sim 9.3 \text{ \AA}$, is approximately same as $d(100)$

of hornblende ($\sim 9.3 \text{ \AA}$). Talc layers could be topotactically coherent with the I-beam units of tremolite at the (010) interface as a result of such dimensional similarity, and the hydration reaction progressed along (100) of tremolite to result in (010) and (100) interfaces (Figs. 6a and 6b). Irregular boundaries at some part of tremolite crystals area (Figs. 6c and 6d) is apparently indicative of differential reaction rate of talc along (100) of tremolite. However, (001) of the replacing talc layers is consistently parallel to [001] of tremolite, suggesting a structural control of tremolite for the formation of talc layers. Such structural relationship is similar to many HRTEM studies of chain silicates that are prone to hydration reactions, resulting in sheet silicates as well as intergrown wide-chain pyriboles (e.g., Smith, 1977; Veblen and Buseck, 1979, 1980, 1981; Nakajima and Ribbe, 1981; Akai, 1982; Sharp and Buseck,

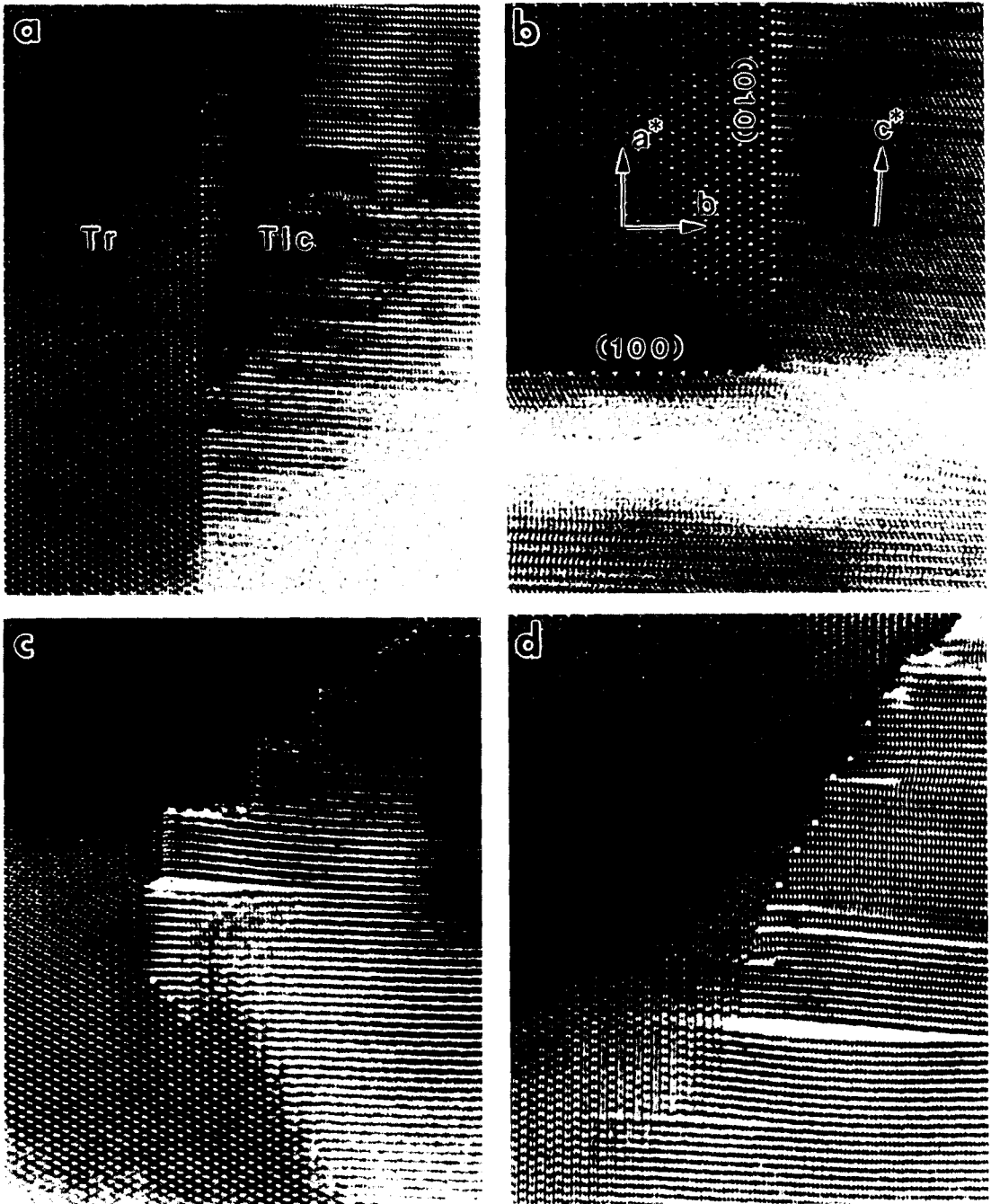


Fig. 6. HRTEM images showing various types of interfaces between tremolite and talc. The interface between tremolite and talc are well defined by (010) of tremolite (a), and (100) of tremolite could constitute well-defined boundary with talc (b). The boundaries between tremolite and talc could be somewhat irregular and diffuse, and the lattice fringes of talc partly overlap with tremolite at the interfaces (c and d).

1988; Ahn and Buseck, 1991; Livi and Veblen, 1992); solid-state reaction of chain silicates commonly proceeds along (010) to produce sheet silicates and intergrown wide-chain pyriboles.

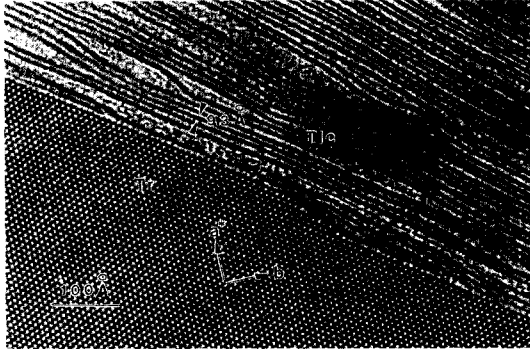


Fig. 7. Talc could develop along (110) of tremolite. The interfaces are partly serrated, and (001) of talc layers are parallel to the (110) interface of tremolite.

Talc associated with tremolite was developed mostly along the (010) and (100) boundaries of tremolite, but some talc could form along (110) of tremolite (Fig. 7). In that case, talc layers are not continuous along (010) of tremolite, but (001) of talc layers are in general parallel to (110) of tremolite indicating that talc layers of that orientation did not form topotactically from tremolite through solid-state mechanism. Such texture indicates that part of talc formed through a dissolution-precipitation mechanism at the Dongyang talc deposit.

Wide-chain pyriboles are frequently observed within tremolite, indicating that wide-chain pyriboles also replace tremolite (Figs. 3 and 9). Chain-width defects in tremolite consist primarily of triple-chain structures together with minor quintuple-chain slabs. The intergrown pyriboles are apparently produced during hydration reaction of tremolite, although a possibility that

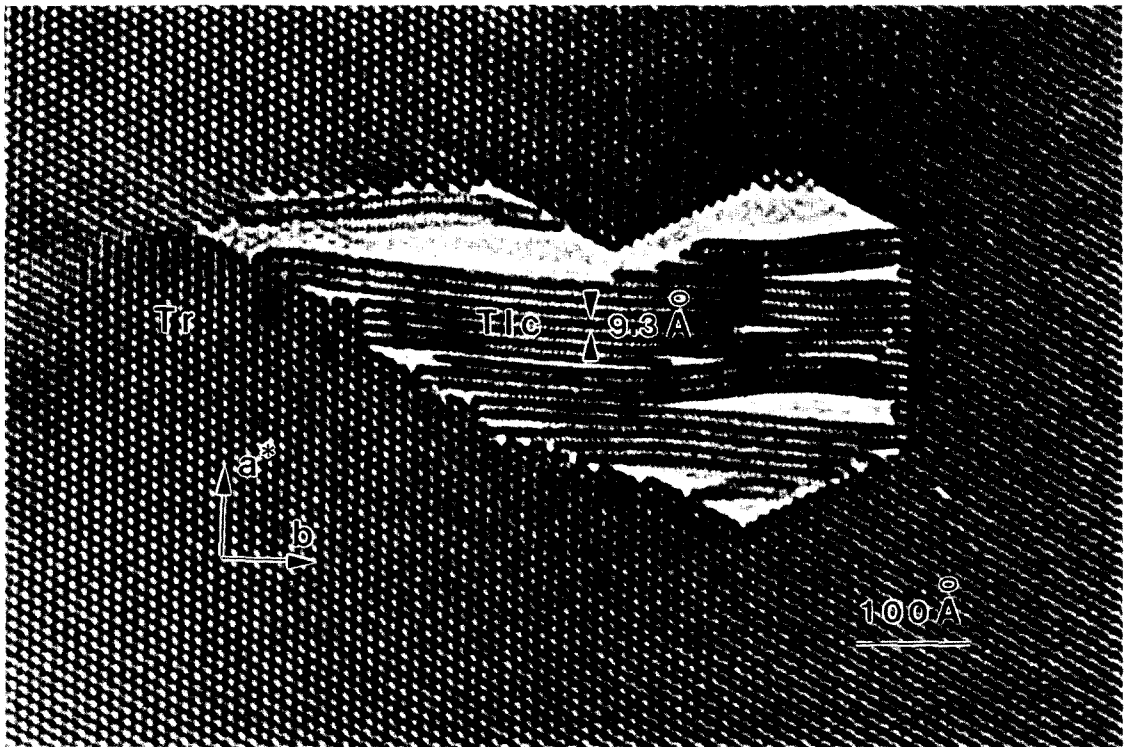


Fig. 8. HRTEM image showing a hole within tremolite crystal. The holes is filled with "wormy-shaped" talc layers that are partly extended from the tremolite "I-beam" units.

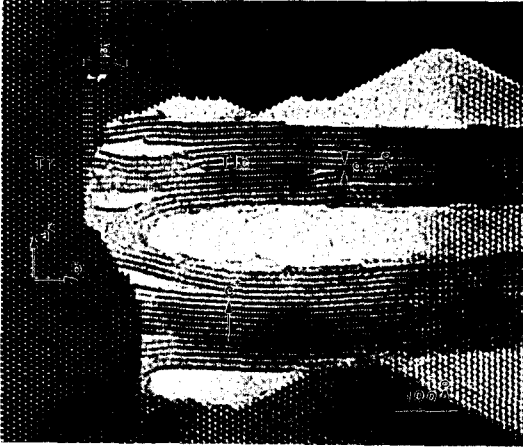
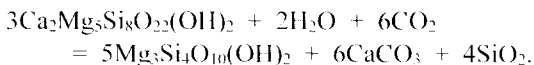


Fig. 9. Wide-chain structure units are associated with the talc developed within a hole. A quintuple chain terminates at one end and takes a step to continue to another quintuple chain. The integer 5 marked on the image indicates intergrown quintuple-chain slabs.

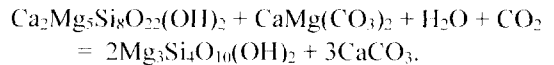
some could have been formed as growth defects during primary crystallization of tremolite. Intergrowth of wide-chain pyriboles within chain silicates was reported to occur during retrograde metamorphism and hydrothermal alteration (Veblen and Buseck, 1979, 1980, 1981; Akai, 1982; Sharp and Buseck, 1988). The volume of such intergrown wide-chain pyriboles are negligible in the investigated specimens, and talc is the primary alteration product of tremolite.

Carbonate minerals are also associated with tremolite and talc; fine-grained calcite occurs along with talc within tremolite crystals (Fig. 1), indicating that calcite formed along with talc from tremolite by the reaction suggested by Park *et al.* (1995),



Furthermore, the replacement texture of dolomite by calcite (Fig. 2) indicates that the tremolite-to-talc reaction could be accompanied with dolomite-to-calcite reaction. Park *et al.*

(1995) also observed that talc occurs with fine calcite grains and forms rim around the tremolite crystal outline which is in contact with dolomite, and they suggested a talc formation reaction;



Their observation is in agreement with our BSE image data. The tremolite-to-talc reaction results in remnant Ca, and that Ca was used to form calcite replacing dolomite. However, calcite formation from dolomite releases Mg that can be utilized to crystallize talc, in that formation of talc from tremolite requires extra Mg.

HRTEM images reveal that tremolite contains holes of various size that are partially filled with talc (Figs. 8 and 9). The holes are apparently extended along the chain direction [001], and some empty gaps are existent inside the holes. Such empty spaces can serve as large tunnels for transport of fluids, providing conduits for fluids and thus facilitating the rate of tremolite-to-talc reaction in tremolite crystals. Although alteration of tremolite progressed mainly along cleavages and fractures, development of holes inside tremolite crystals may also have played an important role in expediting the alteration of tremolite to talc.

Park *et al.* (1995) suggested two types of tales, namely leafy talc and microcrystalline talc, based on the crystallization stage and textures; leafy talc and tremolite formed by the reaction of dolomite and the fluids at an early stage, and microcrystalline talc formed by the reaction of early formed tremolite by fluids. Most talc associated with tremolite in this study apparently corresponds to microcrystalline talc described by Park *et al.* (1995). Extensive alteration of tremolite to talc as reported in this study suggests that the formation of talc directly from tremolite was also responsible for significant part of talc at the Dongyang talc deposit.

Conclusions

HRTEM observation of tremolites from Dongyang talc deposit shows that tremolite crystals remained within talc contain talc and intergrown wide-chain pyriboles. Talc is the primary alteration product of tremolite, and triple-chain pyriboles occur only in minor amount. Tremolite and talc commonly show well-defined interfaces along (010) and (100) of tremolite. Talc layers are continuous to (100) of tremolite, and the interfaces between tremolite and talc appear to be topotactic and coherent in HRTEM images. HRTEM data indicate that talc layers formed directly from tremolite by a hydration reaction and that some talc formed through dissolution-precipitation mechanism along (110) of tremolite. Replacement of dolomite by calcite indicates that carbonate minerals are also associated with the tremolite-to-talc reaction; the formation of talc from tremolite resulted in remnant Ca, which was eventually utilized in the replacement of dolomite by calcite, and Mg released by dolomite during reaction to calcite aided tremolite to react to talc. Most talc associated with tremolite in this study is apparently microcrystalline talc described by Park *et al.* (1995), and extensive alteration of tremolite to talc suggests that the tremolite-to-talc reaction also have resulted in substantial amount of talc of the Dongyang talc deposit.

Acknowledgments

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