# Biotite chloritization by interlayer brucitization as seen by HRTEM

JUAN OLIVES BAños and MARC AMOURIC

CRMC2, Campus de Luminy, 13288 Marseille cedex 9, France

# Abstract

High-resolution transmission electron microscopy provides direct observation of the elementary talc-like and brucite-like layers that compose the structures of micas and chlorites, and precise information on the internal or stacking structure of these layers. Observations of metamorphic biotites partially altered into chlorite show that chloritization occurs by brucitization of the interlayer levels of biotite (potassium planes). This process leads to all possible (ordered and disordered) interlayered biotite–chlorite structures. The ordered 1 biotite–1 chlorite structure has been frequently observed. The shifts between successive tetrahedral sheets of talc layers ("talc-staggers" and "brucite-staggers") can be obtained from high-resolution images taken at optimal observation conditions.

#### Introduction

Interlayered structures of phyllosilicates can be observed directly with transmission electron microscopy (TEM) (McKee and Buseck, 1978; Page and Wenk, 1979; Knipe, 1981). Isolated chlorite layers interstratified in biotite have been shown by Iijima and Zhu (1982) and Veblen and Ferry (1983) and with high-resolution TEM (HRTEM), Veblen and Buseck (1980, 1981) and Veblen (1983) have distinguished the talc-like and brucite-like sheets that compose a chlorite layer and observed brucite-like sheets interlayered in talc and wonesite. This paper presents HRTEM observations of metamorphic biotites (Bormes gneisses, Maures massif, France) partially altered into chlorite.

The Bormes gneiss is an original intrusive granite affected by Barrovian progressive metamorphism (Tempier et al., 1980). The gneiss contains quartz and feldspar layers separated by thin layers of muscovite and biotite. Chlorite is not stable in this zone of metamorphism, but the biotites show some small localized regions altered to chlorite. In a preceding study (Olives et al., 1983), we have described the brucitization of an interlayer level of biotite (equivalent to a unit layer chloritization) and shown that this chemical transformation is strongly favored by pre-existing deformation defects. The observation of undeformed biotites, which are partially chloritized, has revealed that this elementary mechanism may act in many interlayer levels (even if deformation defects are absent) and is responsible for biotite alteration to chlorite. We describe here the successive states of the chloritization process and the structural data that are given by HRTEM images. The samples were prepared from conventional petrographic thin sections. Selected zones were thinned in an argon ion miller (for more details on the technique, see Olives et al., 1983). Observations were made with a JEM 100C microscope (accelerating voltage 100 kV, spherical aberration coefficient 1.7 mm, objective aperture 35  $\mu$ m). Images were taken at -1200Å defocus, the optimum value for biotite (Amouric et al., 1981).

## **Biotite chloritization**

We have observed, as the earliest states of the chloritization process, isolated brucite-like partial layers that terminate on an interlayer level of the biotite (Fig. 1a, 1c, lower part; Olives et al., 1983, Fig. 3). The images show clearly that the chloritization mechanism is basically the replacement of a potassium plane by a brucite-like sheet; i.e., brucitization of an interlayer level of the biotite. This mechanism produces a single chlorite layer consisting of the brucite sheet and either of the two adjacent talc-like layers.

The upper part of Figure 1a shows a more chloritized zone; however, chloritization is not complete since brucite sheets alternate with untransformed interlayer levels of the mica. Thus, a structure consisting of one single layer of biotite alternating with one single layer of chlorite is locally formed. Such ordered structures have been frequently observed. In Figure 1b, the structural unit 1 biotite layer-1 chlorite layer is repeated fifteen times, with only one fault consisting of two adjacent chlorite layers. In fact, all intermediate (ordered and disordered) states between untransformed biotite, 1 biotite-1 chlorite structure and completely chloritized zones, can be observed, as illustrated in Figure 1c. The last state before complete chloritization is shown in Figure 1d: only one partial interlayer level of primitive biotite has not been brucitized.

# **High-resolution imaging**

The two tetrahedral sheets T of a talc layer T-O-T are shifted relative to one another by a "layer stagger"  $\vec{a}_i/3$ 



Fig. 1. Different states of biotite chloritization. I (thin white fringes) denotes interlayer levels of biotite (potassium planes) and B brucite-like sheets. Talc-like layers (not indicated on the images) are situated between successive I or B levels. Each I level corresponds to a single biotite layer and each B sheet to a single chlorite layer. (a) Early state of biotite chloritization. (b) Intermediate state showing ordered 1 biotite–1 chlorite structure. (c) Intermediate disordered state. (d) Almost completely chloritized region.

(Smith and Yoder, 1956), which we shall call briefly "talc-stagger" ( $\vec{a}_i$  denotes any of the six vectors joining two neighboring centers of hexagons of tetrahedra). In chlorite, the stacking structure of talc and brucite layers (Brown and Bailey, 1962) shows that the two tetrahedral sheets (of talc layers) separated by a brucite sheet may also be shifted: this stagger, which will be called briefly "brucite-stagger", may be equal to  $\vec{a}_i/3$  (as for talc-staggers) but also to  $\vec{0}$  and to  $\vec{a}_i/3 + \vec{a}_j/3$  with  $(\vec{a}_i,\vec{a}_j)^3 = 60^\circ$  (Olives et al., 1983, Fig. 1c).

In micas, when the electron beam is parallel to an  $\vec{a}_i$  direction, high-resolution images show lines of white dots of 4.5Å spacing, and two successive dotted lines may be shifted by 0 or  $\pm 1.5$ Å (Iijima and Buseck, 1978; Amouric et al., 1978). These observed shifts are consistent with talc-staggers projected onto the image plane. Iijima and Buseck (1978) and Amouric et al. (1978) proposed a direct structural interpretation of the white spots, as corresponding to the "tunnels" of low electron density situated between the rows of potassium ions. Calculated images have proved that white spots correspond to tunnels only under precise observation conditions (Amouric et al., 1981).

In a high-resolution image taken at optimal observation conditions (Fig. 2), two successive interlayer levels of biotite have been brucitized forming two layers of chlorite. In biotite, as noted above, the shifts between successive dotted lines correspond to the projected talc-staggers, being equal to 0 for the two upper and lower biotite layers of Figure 2. In chlorite, the shift between dotted lines is equal to 1.5Å for the two brucite sheets, and to 0



Fig. 2. High-resolution image of biotite with two interstratified chlorite layers. Same notations as in Figure 1. Talc-like layers are indicated by Ta. Shifts of 1.5Å between dotted lines are indicated by arrows.

for the talc layer situated between them. Since talcstaggers and brucite-staggers have identical projections of 0 or  $\pm 1.5$ Å when viewed along  $\vec{a}_i$ , the preceding image shifts correspond very probably to projected talc-staggers and brucite-staggers. Two particular talc layers are limited on one side by a potassium plane and on the other side by a brucite sheet; the shift between dotted lines for these two layers is equal to 1.5Å. Here also the shift probably corresponds to the projected talc-stagger.

### Conclusions

Biotite chloritization is a chemical transformation that occurs here in the interlayer levels of biotite ("interlayer brucitization"). A different mechanism of chloritization ("talc brucitization") has been recently observed by Veblen and Ferry (1983). In our samples, the elementary mechanism is the replacement of a plane of potassium ions by a brucite-like sheet (this process being favored in the interlayer levels where partial slip or cleavage have occurred; Olives et al., 1983). It produces chlorite layers interstratified in biotite and all intermediate states between biotite and chlorite can be observed. Ordered structures, such as the 1 biotite-1 chlorite structure, may be locally formed. In high-resolution "dotted" images taken at optimal observation conditions, shifts between dotted lines represent the projected staggers between successive tetrahedral sheets ("talc-staggers" and "brucite-staggers").

#### References

- Amouric, M., Baronnet, A., and Finck, C. (1978) Polytypisme et désordre dans les micas dioctaédriques synthétiques. Etude par imagerie de réseau. Material Research Bulletin, 13, 627– 634.
- Amouric, M., Mercuriot, G., and Baronnet, A. (1981) On computed and observed HRTEM images of perfect mica polytypes. Bulletin de Minéralogie, 104, 298-313.
- Brown, B. E. and Bailey, S. W. (1962) Chlorite polytypisme: I. Regular and semi-random one-layer structures. American Mineralogist, 47, 819–850.

- Iijima, S. and Buseck, P. R. (1978) Experimental study of disordered mica structure by HRTEM. Acta Crystallographica, A 34, 709–719.
- Iijima, S. and Zhu, J. (1982) Electron microscopy of a muscovite-biotite interface. American Mineralogist, 67, 1195-1205.
- Knipe, R. J. (1981) The interaction of deformation and metamorphism in slates. Tectonophysics, 78, 249–272.
- McKee, T. R. and Buseck, P. R. (1978) HRTEM observation of stacking and ordered interstratification in rectorite. In Strugess, Ed., Electron Microscopy, 1, Microscopical Society of Canada, Toronto, 272–273.
- Olives, J., Amouric, M., de Fouquet, C. and Baronnet, A. (1983). Interlayering and interlayer slip in biotite as seen by HRTEM. American Mineralogist, 68, 754–758.
- Page, R. H. and Wenk, H. R. (1979) Phyllosilicate alteration of plagioclase studied by transmission electron microscopy. Geology, 7, 393–397.
- Smith, J. V. and Yoder, H. S. (1956) Experimental and theoretical studies of the mica polymorphs. Mineralogical Magazine, 31, 209-235.
- Tempier, C., Bronner, G., Gueirard, S., and Lécorché, J. P. (1980) Signification géologique des gneiss de Bormes. Chronologie des évènements dans le massif des Maures (Var).8<sup>e<sup>'me</sup></sup> Réunion Annuelle des Sciences dè la Terre, Marseille, Société Géologique de France, 341.
- Veblen, D. R. (1983) Microstructures and mixed layering in intergrown wonesite, chlorite, talc, biotite and kaolinite. American Mineralogist, 68, 566–580.
- Veblen, D. R. and Buseck, P. R. (1980) Microstructures and reaction mechanisms in biopyriboles. American Mineralogist, 65, 599–623.
- Veblen, D. R. and Buseck, P. R. (1981) Hydrous pyriboles and sheet silicates in pyroxenes and uralites:intergrowth microstructures and reaction mechanisms. American Mineralogist, 66, 1107–1134.
- Veblen, D. R. and Ferry, J. M. (1983) A TEM study of the biotite-chlorite reaction and comparison with petrologic observations. American Mineralogist, 68, 1160-1168.

Manuscript received, July 7, 1983; accepted for publication, March 7, 1984.