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NOTE

ALTERNATION OF CLINO- AND ORTHOCHRYSOTILE IN A SINGLE FIBER AS REVEALED BY HIGH-RESOLUTION ELECTRON MICROSCOPY

Key Words—Asbestos, Alternate structures, Chrysotile, Defect, Fiber, Transmission electron microscopy.

Direct observations of the atomic arrangements of crystals by high-resolution electron microscopy (HRTEM) have been reported frequently in the past fifteen years. Most of these observations, however, were made on materials that are stable in the electron beam (e.g., metals, semiconductors, oxides). For materials sensitive to the electron beam, highly refined techniques and special devices are required (Yada, 1967, 1971; Yada and Iishi, 1977; Fujiyoshi *et al.*, 1980), and, thus, studies of such materials have been limited.

In previous studies of the microstructures of serpentines, especially chrysotile, a material that is very sensitive to electron beam irradiation, lattice defects such as dislocation (Yada, 1967), twinning (Yada and Iishi, 1977), and many abnormal growth patterns (Yada, 1971) were observed. In addition, the atomic arrangement of the clino and ortho structural varieties of chrysotile determined by X-ray crystal-structure analysis was recently verified by atomic resolution electron microscopy where the optical filtering method and a modified model for the computer simulation of the rolled structure of chrysotile were employed (Yada, 1979; Yada and Tanji, 1980). Image processing is essential for such a beam sensitive material, and the confirmation of the structural features by computer simulation is necessary.

Chrysotile fibers usually occur in the order clino > ortho > para, although the actual frequency of their occurrence varies from locality to locality. No information, however, as to whether or not these structures coexist within an individual fiber of chrysotile is available. In the present paper, the evidence for the alter-

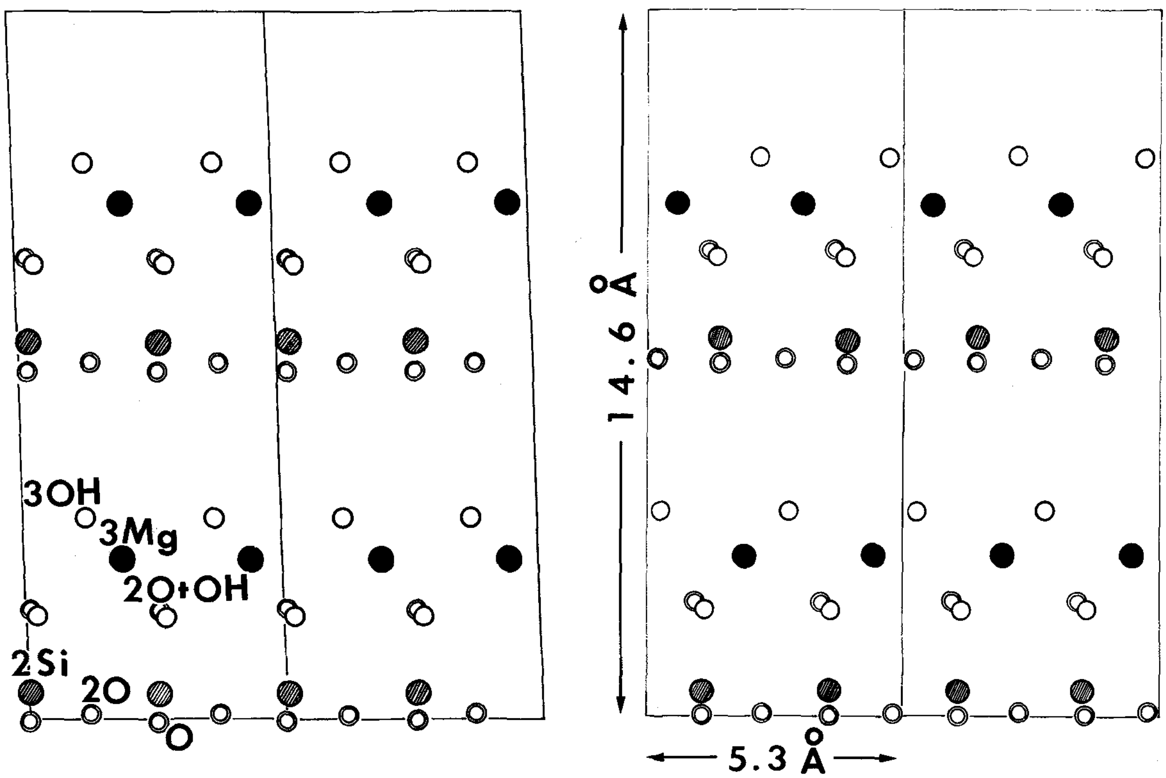

Figure 1. Atomic positions of clinochrysotile (left) and orthochrysotile (right) projected along the (010) direction (after Whittaker, 1956a, 1956b).

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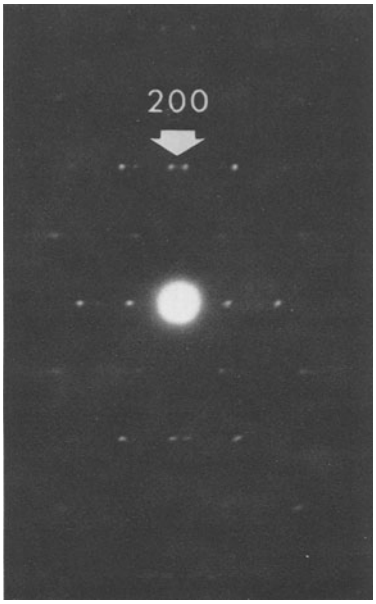

Figure 2. Typical electron diffraction pattern of clinochrysotile. (Coalinga mine, California.)

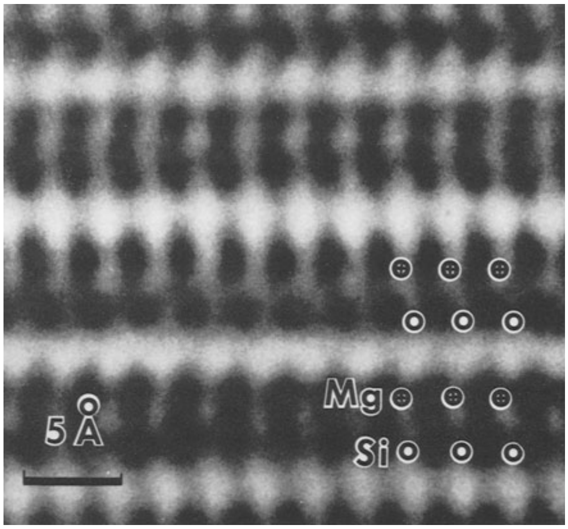

Figure 3. An example of the images of clinochrysotile processed optically with a noise filter. (Jeffry mine, Asbestos, Quebec.)

nation of clino and ortho structures is reported on the basis of the optically processed image of a single chrysotile fiber.

EXPERIMENTAL

The experiments were performed on chrysotile fibers from the Jeffry mine, Asbestos, Quebec, and the Coalinga mine, near New Idria, California. The (010) projection of the atomic positions of clinochrysotile (β = 93.6°) and orthochrysotile (β = 90.0°), as determined by Whittaker (1956a, 1956b) are shown in Figures 1a and 1b, respectively.

Electron microscopes used were the HU-11B (100 kV) and JEM-200CX (200 kV) instruments. The water-suspended chrysotile fibers were mounted on holey carbon films; most were laid horizontally, so that the electron beam was nearly parallel to b-axis of the fiber. Optical noise filtering for periodic objects (O'Neil, 1956; Klug and De Rosier, 1966) was employed to enhance the signal to noise ratio of the image. This technique has what amounts to an averaging effect which acts as if it was due to the multiple superpositions of the object picture with periodic shifts of position in photographic printing (Tanji *et al.*, 1982). The effective range is inversely proportional to the diameter of holes on the filter. Therefore, the filter for the image which has the periodic structure in only one direction may be constructed with the slits lying perpendicular to that direction (Yada and Tanji, 1980). If the object contains a defect in the periodic structure, the width of slits must be somewhat wider than usual in order that the averaging effect does not suppress the contrast of the defect.

A modified commercial diffractometer was used for the noise filtering, and two kinds of filters were tried to compare their filtering effects. One filter was a brass plate with mechanically drilled slits; the other was a highly darkened photographic film with transparent slits. Images processed with both types of filters showed well-defined features in good accordance with each other. In general, film filters were used because of the ease of their preparation.

RESULTS AND DISCUSSION

One of the main features of the electron diffraction pattern of clinochrysotile was the splitting of 200 spots (Figure 2), which suggests that the parameter β deviates 3–3.5° from a right angle. It should be noted that both spots had the same intensity. The reason for the split was discussed by Yada (1967) and was easily confirmed with high resolution images and/or their optical diffraction patterns. The selected-area, optical microdiffraction pattern from a high-resolution micrograph (Tanji and Hashimoto, 1978) was confined to one side of the fiber giving only one of the two split 200 spots (cf. Figure 4b) in contrast to the electron diffraction pattern (Figure 2) where diffractions from both sides of a clino-type fiber were recorded simultaneously.

Figure 3 shows a typical image of clinochrysotile taken with a 100-kV electron microscope. Computer simulation of the image contrast showed that the intensity maxima or minima did not always coincide with the correct positions of Mg and Si atoms, because these two elements are not significantly heavier than

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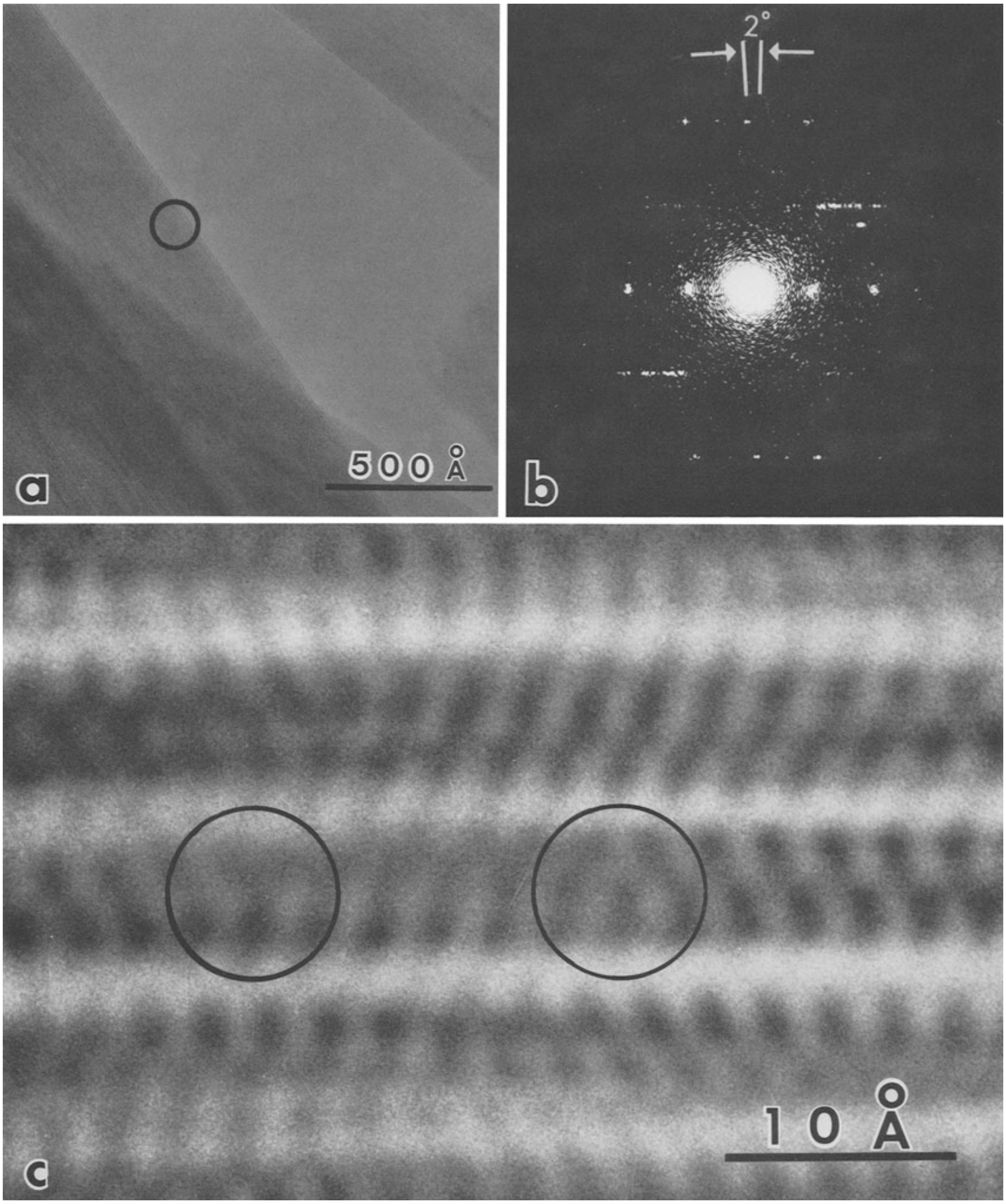

Figure 4. (a) Electron micrograph of chrysotile fibers; (b) Optical diffraction pattern of the encircled area in (a); (c) Optically processed image of the same area as (b), but shown with a right-angle rotation. The lower layer is on the core side of the fiber. (Jeffry mine, Asbestos, Quebec.)

their surroundings and Scherzer's optimum was not valid for such fine structures. It has been shown, however, that the arrangement of intensity maxima and minima reflects that of true atoms which are indicated commonly by the positions of Si and Mg atoms (Yada and Tanji, 1980).

Figure 4a shows the image of a chrysotile fiber taken with a 200 kV electron microscope. Figure 4b is an optical diffraction pattern of the encircled area in Figure 4a, and Figure 4c is the corresponding processed image. Although the inclination of 200 spots (~2°) in the diffraction pattern seems to suggest that the fiber

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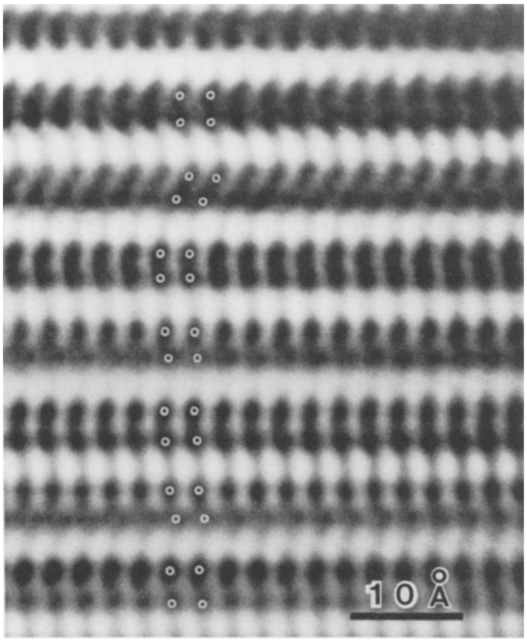

Figure 5. Interlayer alternation illustrated by the arrayed marks which are referred to the arrangement of Si and Mg atoms. The clino-type layer alternates with an ortho-type layer (the upper part in the picture). The lower layer is on the core side of the fiber. (Jeffry mine, Asbestos, Quebec.)

is composed of the clino-type structure (see Figures 2 and 3), in the circles of Figure 4c the inclination of the pairs of dark spots is inverted within a single 7.3-Å layer. From the relationship of the positions of Mg and Si atoms in the adjacent layers, it seems that the clino type in some areas of a 7.3-Å layer changes to the ortho type in other areas of the same layer. The encircled areas in Figure 4c contain transitions between the two structures and represent intralayer transitions. Figure 5 shows another example of a clino to ortho transition within a single fiber. Here, an interlayer change of the stacking sequence can be seen. The image was obtained with an 100 kV electron microscope using an image-processing technique.

From these results, the alternation of clino- and ortho-type structures within a single chrysotile fiber can be shown. Such localized transitions cannot be detected with electron microdiffraction, because their detection requires that the diameter of the diffraction area be limited to a few tens of Ångstrom units; chrysotile is too sensitive to the electron beam to withstand the long exposure required. More experiments and computer simulations are needed to determine the details of the atomic arrangements at the structural transitions, especially at the intralayer transition.

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