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TRANSMISSION ELECTRON MICROSCOPY OF NATIVE COPPER INCLUSIONS IN ILLITE

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INTRODUCTION

The occurrence of anomalous copper concentrations in biotites from rocks associated with porphyry copper deposits has long been of interest (Banks 1974; Rehrig and McKinney 1976; Hendry et al. 1981). Submicrometer particles of native copper were discovered in biotites through transmission electron microscope (TEM) and electron microprobe investigations (Ilton and Veblen 1988, 1993); such inclusions were produced experimentally using copper sulfate solutions at atmospheric temperature and pressure (Ilton et al. 1992). Since native copper inclusions are found only in altered sheet silicates, Ilton and Veblen (1988, 1993) concluded that the inclusions formed during weathering rather than during the hydrothermal events. Their findings suggest that sheet silicates act as copper sinks rather than as sources for primary mineralization (Titley 1988).

Since sheet silicates other than biotite and phlogopite are also associated with copper deposits, a remaining question is whether only trioctahedral micas can be the hosts of copper inclusions. In the present study, we report the occurrence of submicrometer copper inclusions in weathered illites. Such inclusions suggest that a variety of sheet silicates other than trioctahedral micas can accommodate copper in their interlayers and could be useful indicators of Cu enrichment.

EXPERIMENTAL METHODS

The specimen investigated in this study is a slaty rock collected from an outcrop located north of the Picacho Mountains, Pinal County, Arizona. The sample is highly weathered, and malachite and azurite are scattered around the sampled area. To obtain optimum orientation for imaging (001) lattice fringes of the sheet silicates, thin sections were prepared by cutting perpendicular to the cleavage direction. Selected areas were investigated using a JEOL 200CX 200-kV TEM with a top-entry stage having tilting angles of $\pm 12°$ and spherical aberration coefficient (Cs) of 1.2 mm.

RESULTS AND DISCUSSION

TEM observation reveals the widespread occurrence of partially expanded silicate layers and inclusions within the illite crystals (Figure 1); the inclusions have flattened shapes and show darker contrast than the surrounding silicate layers. The inclusions only occur in the areas where the 10-Å layers gradually expand along the layer direction; they do not occur in unaltered illite layers that show regular 10-Å lattice fringes. Most inclusions in the illite are less than 30 Å in thickness and 300 Å in length (Figure 1). The inclusions cause gentle bending of the surrounding silicate layers.

The inclusions found in this study are similar to those within Cu-rich biotite crystals reported by Ilton and Veblen (1988, 1993), although their dimensions are, in general, smaller than those in the biotites. They identified them as native copper particles based on electron diffraction data. High-resolution TEM images of inclusions show lattice fringes with approximately 2-Å spacings (Figure 2); these match the $d(111)$ value of native copper, so we conclude that these, like the inclusions in biotite, are native copper.

Mica layers are partially frayed as a result of the increased number of expanded layers (Figure 3). The widths of the interlayer regions change gradually along the layer directions, commonly resulting in lenticular area. The separated areas with brighter contrast correspond to widened interlayer regions; TEM image simulations showed that interlayer regions of mica with low or no electron density have bright contrast areas at favorable defocus conditions (Amouric et al. 1981; Ahn and Buseck 1989, 1990; Guthrie and Veblen 1989).

Numerous copper inclusions of various dimensions are located in the interlayer regions, and silicate layers become gently wavy around the inclusions (Figures 1 and 3). It has been known that weathering of dioctahedral micas also produces expandable sheet silicates such as smectite or vermiculite (Jackson et al. 1952; Churchman 1980; Jiang et al. 1990). Our specimens were collected from weathered outcrops, and the lack of sulfide minerals suggests that the expanded layers formed by weathering rather than hydrothermal alteration. In addition, the occurrence of copper inclusions exclusively within the expanded interlayer regions

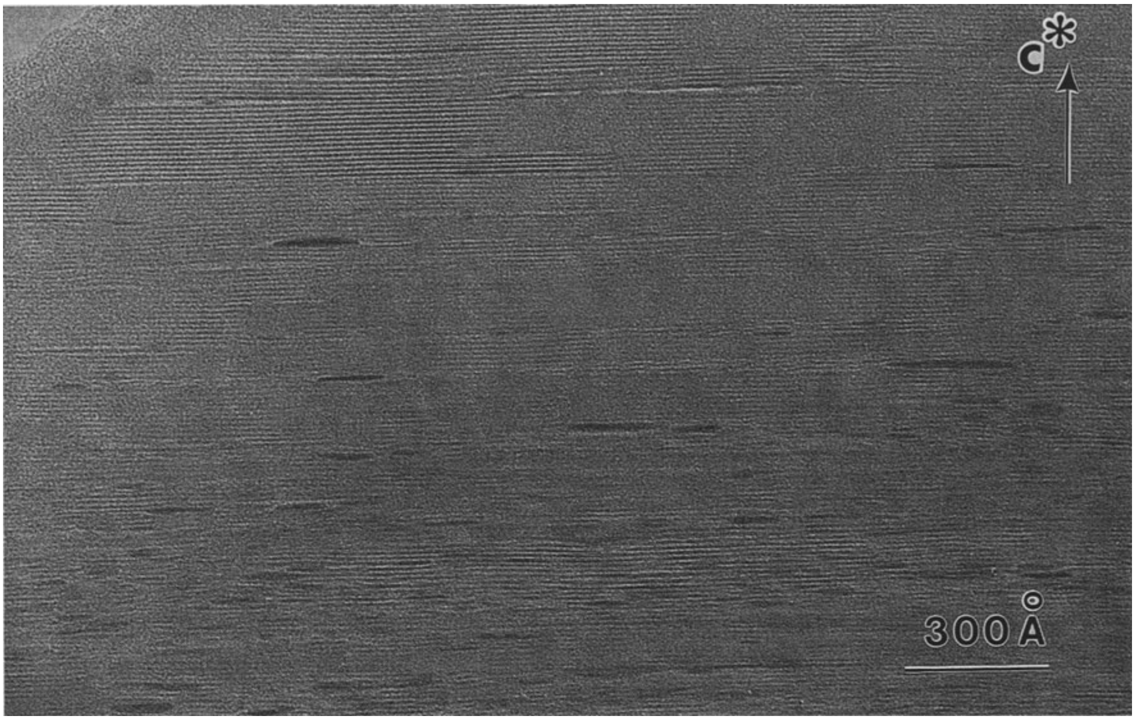

Figure 1. TEM image showing the Cu inclusions scattered in partially altered illite.

suggests that the origin of the copper inclusions is related to the weathering of the micas (Ilton and Veblen 1988, 1993). Cu-rich solutions presumably penetrated the expanded interlayers and resulted in precipitation of the copper inclusions.

Ilton and Veblen (1993) proposed that the introduction of Cu^{2+} into interlayers is related to the release of K^{+} in the micas and that precipitation of Cu^{2+} in the interlayers as native copper is accompanied by the oxidation of Fe^{2+} to Fe^{3+} in the octahedral sites. However, dioctahedral micas contain far less Fe^{2+} than do trioctahedral micas; the oxidation of octahedral iron can only partly account for the precipitation of native copper in dioctahedral micas. Weathering of micas accompanies the decrease of layer charges by leaching of tetrahedral Al and replacement by Si (Sridhar and Jackson 1974), and leaching of tetrahedral Al as well as oxidation of Fe may have played important roles in decreasing the layer charge and precipitation of copper in the illite.

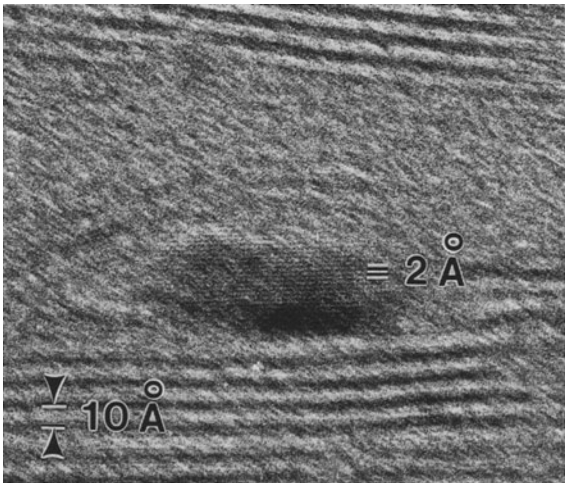

Figure 2. TEM image of a Cu inclusion exhibiting approximately 2-Å lattice fringes.

Gaps where copper inclusions are clearly visible in TEM images are wider than those where inclusions are absent (Figures 1 and 3). The elongation direction of inclusions, (111) of native copper, is everywhere parallel to (001) of mica (Figure 2), indicating that copper atoms were precipitated along the interlayers. Growth of the copper inclusions across (001) of silicate layers apparently caused mechanical separation of interlayers beyond that which occurred during weathering.

The association of inclusions with expanded silicate layers and lack of copper sulfide minerals suggest that copper was introduced during or after initial weathering rather than during primary crystallization of illite. Our observations are consistent with the conclusion of Ilton and Veblen (1988, 1993), who suggested that the occurrence of copper inclusions in micas is related to enriching of copper during weathering rather than during magmatic hydrothermal processes.

Dioctahedral micas apparently contain smaller inclusions than triocthedral micas, and copper inclusions

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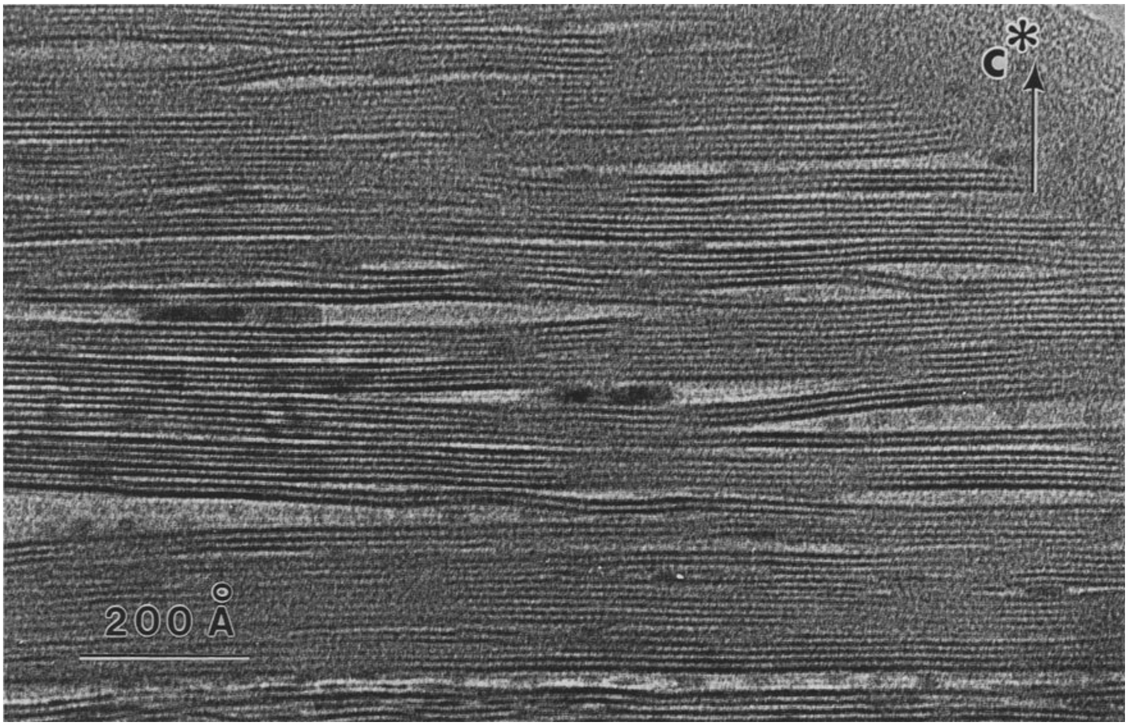

Figure 3. TEM image of highly weathered illite showing partially frayed mica layers and Cu inclusions.

may be precipitated in dioctahedral micas only in highly weathered and oxidized environments. The occurrence of copper inclusions in dioctahedral as well as trioctahedral micas suggests that copper can be precipitated in the interlayers of many different sheet silicate minerals.

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