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SCANNING ELECTRON MICROSCOPIC STUDY OF IMOGOLITE FORMATION FROM PLAGIOCLASE

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Abstract—Imogolite occurs as tiny bumps less than 0.05 μm in diameter and about 0.3 μm long on amorphous thin layers on the surface of weathered plagioclase. The bumps grow outward from the surface to form projections, which then develop into a fibrous imogolite. The fibers branch out and grow into widespread networks or thin films which finally cover the entire surface of the plagioclase grain. The fibers are about 0.05 μm in diameter as seen by transmission electron microscopy.

Key Words—Alteration, Fiber, Imogolite, Plagioclase, SEM, Weathering.

INTRODUCTION

Imogolite is a fibrous clay mineral that occurs in young, glassy volcanic ash soils (Yoshinaga, 1970; Yoshinaga *et al.*, 1968, Wada and Matsubara, 1968; Wada *et al.*, 1970, 1972; Honda, 1972; Tazaki, 1971, 1975; Eswaran, 1972; Noishiki and Tazaki, 1975). Although it has previously been shown to form by the alteration of volcanic glass, the present study describes the formation of imogolite from plagioclase grains in volcanic ash. Studies on naturally weathered or artificially altered plagioclase have revealed the formation of various clay minerals, such as halloysite, kaolin minerals, montmorillonite, and gibbsite, but imogolite has not been reported. The weathering of plagioclase evidently is still not fully elucidated.

MATERIALS AND METHODS

Weathered volcanic ash was collected from 80 sites around Mt. Daisen and Mt. Sambe volcanoes in the San-in District, Japan. Allophane, Al-vermiculite, chlorite, halloysite, metahalloysite, kaolinite, and mica-like minerals have been reported as weathering products of these ashes (Kurahayashi, 1972; Tazaki, 1972). Fragile, lusterless plagioclase phenocrysts occur as prismatic grains 1–2 mm in length. Compositional zoning is common and albite-carlsbad twinning occurs. The chemical composition of typical plagioclase grains indicates an An content of 35–39 mole percent, and the plagioclase may be classified as andesine. The $2\theta_{(131)} - 2\theta_{(1\bar{3}1)}$ values of 1.90–1.95 obtained by powder X-ray diffraction indicates that the plagioclase is a high-temperature type.

Partly altered plagioclase grains were separated from the ash samples by hand picking and hydraulic elutriation methods. They were washed with distilled water and air-dried at room temperature. Thin sections were prepared for microprobe analysis. Replicas of various cleavage surfaces were made using replica tape and shadowed with carbon at an angle of 45° within a thickness of 500–1000 Å for examination by transmission electron microscopy (TEM) using a JEM 100C instrument. Scanning electron microscopy (SEM) was carried out with a JSM 50A instrument with an accelerating voltage of 15 kV. The plagioclase grains were mounted on brass stubs and coated first with carbon and then with gold. Selected grains were also examined by non-dispersive X-ray spectroscopy (EMMA) using an EDAX analyzer. Electron probe analyses were made using a JXA 5A instrument at a take-off angle of 40° under the following conditions: 15 kV, 0.02 μa, 3–5 μm beam diameter on ZrO_2. Synthetic wollastonite, Al_2O_3, and Fe_2O_3 were used as standards for Si and Ca, Al, and Fe, respectively; albite and adularia were used as standards for Na and K, respectively.

Imogolite fibers and films on the surface of weathered plagioclase grains were separated by ultrasonic treatment at 400 kHz for 15 min and centrifugation. The broad X-ray powder diffraction reflection at about 14 Å may be ascribed to the presence of imogolite. The 4.8 Å reflection suggests the presence of a small amount of gibbsite. The characteristic fibriform of imogolite can be seen in transmission electron microscopy.

RESULTS

The surface of fresh plagioclase grains is flat and smooth, whereas the surface of weathered plagioclase shows tiny bumps, projections, cracks, and pits which may be ascribed to the formation of various clay minerals.

Imogolite seems to develop first at exposed edges and corners of fractured surfaces (Figure 1A). The projections grow outwards and then connect each other. They branch out and finally grow into widespread networks or thin films (Figure 1B). Ultimately, the films cover the entire surface of the plagioclase grain (Figure 1C). The tiny bumps and projections that formed on the surface of the artificially weathered K-feldspar (shown by Parham [1969] in electron micrographs) are similar to those observed in the present study. The high-magnification, one-step replica of weathered plagioclase showed a

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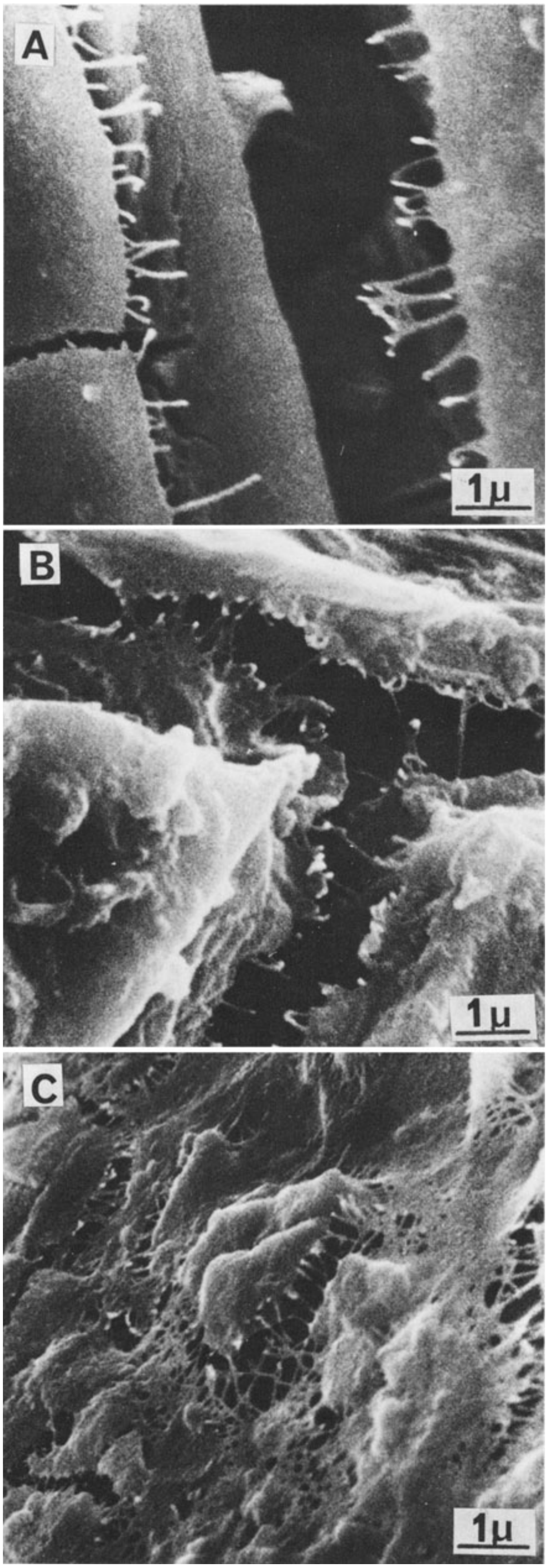

complex network of imogolite fibers. Thick bundles of imogolite threads are clearly visible in Figure 2A. Imogolite networks bridging the hollows in plagioclase grains are shown in Figure 2A. In Figure 2B and C round tips on imogolite threads projected from the network can be seen. The imogolite fiber on the replica is about 0.05 μm in diameter as seen by transmission electron microscopy.

Generally, the micromorphology of the flat plagioclase surface can be investigated in more detail by the TEM replica method than by the SEM. Dispersed imogolite fibers appear to be more wispy under the TEM observation (0.01–0.03 μm) than under the SEM observation (Yoshinaga and Aomine, 1962). The imogolite fibers range from 0.05 to 0.1 μm in diameter under the SEM observation. Fine allophane particles (0.02 μm in average diameter) accompanied by imogolite fibers were also observed in the same sample. The results of semiquantitative elemental analyses for imogolite by EMMA show that the imogolite fibers on the plagioclase grain contain no CaO or Na_2O. They are composed of Al_2O_3 and SiO_2 only, whereas the plagioclase contains CaO and Na_2O.

DISCUSSION

From electron microscopic observations, imogolite presumably grows from projections on plagioclase surfaces by precipitation from solution. The development of a network of imogolite fibers may be the result of repeated or continuous reactions involving the dissolution of alkali and metalic ions from the feldspar and subsequent precipitation of alumina and silica.

Amorphous thin layers on the surface of weathered plagioclase, which are supposed to be chemically unstable, may promote the formation of imogolite (Tazaki, 1978). In this connection, the coexistence of allophane and gibbsite with imogolite has been noted at different Japanese localities (Yoshinaga, 1970; Wada and Matsubara, 1968; Honda, 1972). Imogolite and gibbsite may therefore form by the reaction of permeating solutions and the amorphous, thin layers on plagioclase surfaces under acidic conditions (pH <5). However, the acidity and the concentration of the dissolved elements in the solution must vary during the course of the dissolution and precipitation. Thus, gibbsite may form during the dissolution process, and imogolite by precipitation.

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Figure 1. SEM photographs of imogolite formed on plagioclase. A. Tiny polypoid projections of imogolite formed from the edges of thin layers. B. Imogolite fibers developing into a network or film. C. Surface of the plagioclase covered with a network of imogolite.

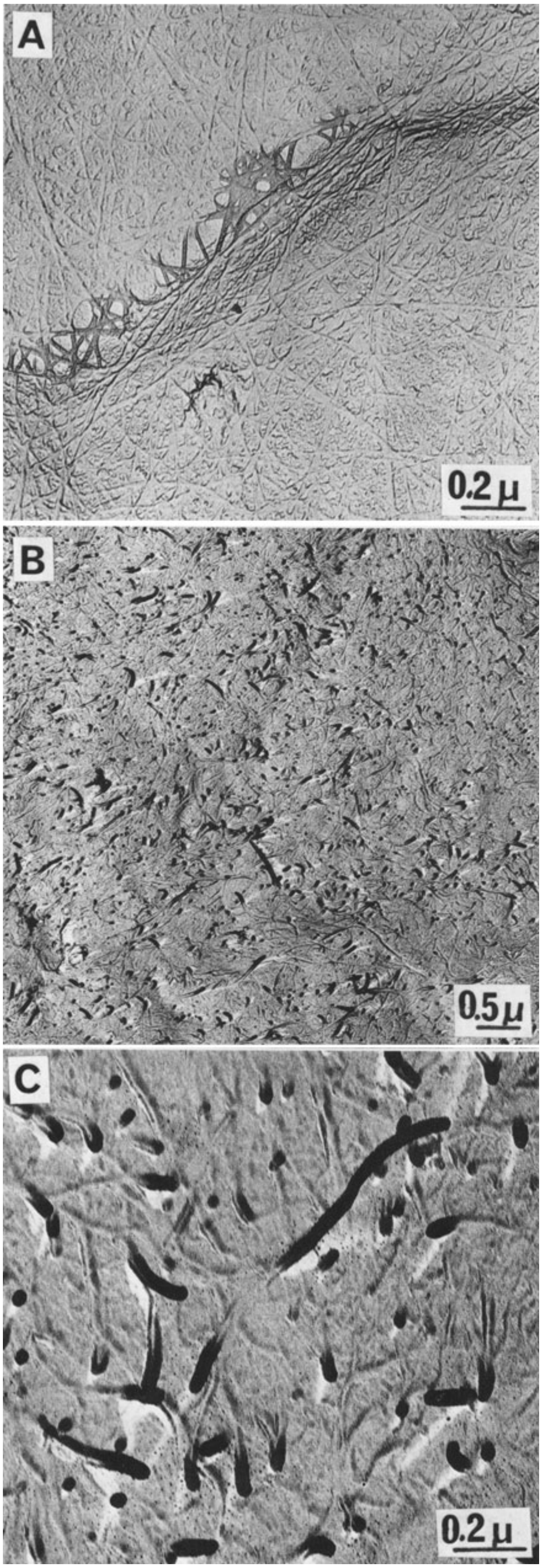

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Figure 2. TEM replica photographs of imogolite formed on plagioclase. A. The flat surface is covered with the imogolite fibers. B. Imogolite fibers standing up on the surface of plagioclase. C. Circular cross sections of imogolite.

Резюме—Имоголит появляется как очень маленькие выпуклости менее чем 0,05 μм в диаметре и около 0,3 μм в длину на аморфных тонких слоях на поверхности выветренного плагиоклаза. Выпуклости растут от наружной поверхности, образуя выступы, которые затем развиваются в волокнистый имоголит. Волокна разветвляются и разрастаются в широко расстилающиеся сети или тонкие пленки, которые в конце концов покрывают всю поверхность зерна плагиоклаза. Наблюдения с помощью трансмиссионного электронного микроскопа показали, что диаметр волокон обычно около 0,05 μм.

Resümee—Imogolit kommt als kleine Blasen vor, welche weniger als 0,05 μm im Durchmesser und ungefähr 0,3 μm in Länge messen und sich auf dünnen, amorphen Schichten auf den Flächen von verwettertem Plagioklas befinden. Die Blasen wachsen nach außen von der Fläche heraus, um Projektionen zu formen, die sich dann zu faserigem Imogolit entwickeln. Die Fasern verzweigen sich und wachsen zu einem ausgedehnten Netz oder dünnen Filmen, welche endlich die gesamte Oberfläche des Plagioklaskerns bedecken. Die Fasern sind ungefähr 0,05 μm im Durchmesser, was mit Rasterelektronenmikroskopie gesehen werden kann.

Résumé—L'imogolite existe comme petites bosses de moins de 0,05 μm de diamètre et d'à peu près 0,3 μm de long sur de fines couches amorphes sur la surface de plagioclase altéré à l'air. Les bosses croissent de la surface vers l'extérieur pour former des projections qui se développent alors en imogolite fibreuse. Les fibres s'étendent et croissent en de larges réseaux ou en films fins qui couvrent finalement la surface entière du grain de plagioclase. Les fibres ont un diamètre d'à peu près 0,05 μm, vus par le microscope électronique par transmission.