THE GEOLOGY OF VERMICULITE OCCURRENCES

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ABSTRACT

Vermiculite occurrences can be divided into two major categories, those having macroscopic and those having microscopic or clay vermiculite. This paper is concerned with the former. The macroscopic type of vermiculite deposits can be subdivided into four categories based on the host rock: (1) ultramafic and mafic; (2) gneiss and schist; (3) carbonate rocks; and (4) granitic rocks. Field and laboratory evidence strongly suggests that most and possibly all macroscopic-type vermiculite deposits formed by the action of supergene solutions on biotite and phlogopite. Vermiculite in mixed-layer association with biotite, phlogopite, and chlorite is likewise believed to be of supergene origin.

INTRODUCTION

Macroscopic and microscopic types of vermiculite deposits differ in some basic aspects. Macroscopic vermiculites are trioctahedral and have a relatively narrow range of cation exchange capacity. Microscopic or clay vermiculites may be either trioctahedral or dioctahedral (Rich and Cook, this volume) and are much more variable in composition and cation exchange capacity, making them difficult in many instances to distinguish from montmorillonite.

Macroscopic vermiculite occurs in four types of host rocks: (1) ultramafic and mafic; (2) gneiss and schist; (3) carbonate rocks; and (4) granitic rocks. In turn each of these has characteristic features. All the major commercial deposits belong to the first category, and the material that is mined is mixed-layer vermiculite–biotite or vermiculite–phlogopite. The ultramafic–mafic type also provides some of the most interesting mineralogic and petrographic relationships. In the gneiss–schist type, the vermiculite occurs as layers in banded metamorphic sequences. In the third category, vermiculite flakes close to the magnesian end member are sometimes found distributed through marbles ranging from calcite to magnesite composition. The fourth category refers to biotite in granitic rocks that has weathered to an expanded or partially expanded alteration product of biotite that puffs when heated in a flame.

A perennial problem in the study of vermiculites is the question of hydrothermal vs. supergene origin. A review of the literature reveals that investigators' opinions are nearly evenly split on this matter. In this paper their evidences are discussed and the arguments favoring a supergene origin

are found to be more convincing. A selected bibliography of reports having bearing on the origin of macroscopic vermiculite is included at the end of this paper.

ULTRAMAFIC AND MAFIC OCCURRENCES

There are striking mineralogic and petrographic similarities among the deposits in this group. These features are epitomized in the occurrence at Day Book, North Carolina (Kulp and Brobst, 1954). A dunite body has been intruded by a pegmatite, and alteration zones exist along the contact between the two. As one passes from the dunite across these zones to the pegmatite, he crosses serpentine, talc, anthophyllite, vermiculite, and finally pegmatite. The vermiculite at Day Book extends to a depth of 40 ft. Below that the material is phlogopite.

In other deposits of this type the same basic pattern can be found with variations. In the dunite bodies east and southwest of Ashville, North Carolina (Hadley, 1949; Hunter and Mattocks, 1936; Mankin and Cassidy, 1960; Murdock and Hunter, 1946; Ross and Shannon, 1926), vermiculite occurs in relationships very similar to those described by Kulp and Brobst. In places some of the zones are missing and in other places the vermiculite occurs in fractures away from the pegmatite intrusives. In still other places the vermiculite occurs along contacts between the dunite and the country rock. In all these deposits, however, the vermiculite apparently is genetically related to the intrusion of the dunite by pegmatite dikes.

At Brinton Quarry, south of West Chester, Pennsylvania (Larsen, 1928; Gordon, 1921; Bassett and Lapham, unpublished investigation), the exposed ultramafic is serpentine. The zones are similar to those reported by Kulp and Brobst (1954), with the exception that chlorite is intimately mixed with the vermiculite in some of the vermiculite zones, and the pegmatite has been identified as a desilicated albitite.

Mixed-layer vermiculite–chlorite occurs in veins and at pegmatite–soapstone contacts in the soapstone–serpentine bodies at the Gregory and Carl Moss properties south of Llano, Texas (Clabaugh and Barnes, 1959; and E. J. Weiss, personal communication). A shaft at the Carl Moss property has revealed vermiculite–chlorite to a depth of 50 ft below the surface.

The major producing vermiculite deposits are at Libby, Montana; Enoree, South Carolina; Tigerville, South Carolina; and Loolekop, Palabora, northeast Transvaal (Bassett, 1959; Buie and Stewart, 1954; Gevers, 1949; Pardee and Larsen, 1929; Schwellnus, 1938). These deposits are pyroxenites which have been intruded by syenite dikes. The "vermiculite" in the Montana and South Carolina deposits is mixed-layer vermiculite–biotite which has been called hydrobiotite by Gruner (1934). The exchangeable cations in the vermiculite layers are magnesium and calcium. The material at Palabora is mixed-layer vermiculite–phlogopite. The "vermiculite" in the pyroxenite bodies does not occur in zones but is disseminated throughout the pyroxenite. In these deposits are remnant masses of unaltered pyroxene and, quite

separate from them, masses of unaltered biotite or phlogopite. Tremolite occurs as veins and along cleavage planes of the pyroxene, biotite, and "vermiculite". Two features of these deposits make them desirable sources of commercial "vermiculite": (1) the "vermiculite" is disseminated and can be mined inexpensively by strip mining methods, and (2) biotite layers in the lattice provide strength to the structure, permitting it to puff before the water vapor can escape from the structure (Bassett, 1959).

At Tigerville, South Carolina, Buie and Stewart (1954) reported that biotite is nearly absent at the surface but is present at a depth of 15–20 ft. At Libby, Montana, mining operations have extended to a depth of 200 ft below the surface, where the ratio of "vermiculite" to biotite is not significantly different from that found at the surface.

At Encampment, Wyoming, amphibolite bodies have been altered to vermiculite. Associated granite and schist have been sericitized. Vermiculite pseudomorphs after amphiboles and garnet are found in the granite and schist (Hagner, 1944; Bassett, unpublished investigation). In places the amphibolite (altered in part to vermiculite) occurs as layers in the gneiss, making the relationships very similar to the gneiss–schist type deposits described in the next section. The Encampment deposits have been worked sporadically for commercial vermiculite, but the yield has been very low.

GNEISS AND SCHIST OCCURRENCES

On U.S. Hwy. 285, 3 miles south of Poncha Springs, near Salida, Colorado, is a road cut exposing contorted gneiss. Vermiculite occurs as dark bands in this gneiss. North of Las Vegas, New Mexico, is a similar occurrence of vermiculite in gneiss. Serpentine bodies near this occurrence also carry vermiculite and may be genetically related to it. The Bush property, north of Llano, Texas, has been described by Clabaugh and Barnes (1957; 1959) and McMillan and Gerhardt (1949). Although the "vermiculite" (mixed-layer vermiculite–biotite) at the Bush property is seemingly a layer in the gneiss, Clabaugh and Barnes have produced convincing evidence that the vermiculite was originally a gabbro sheet that was altered to an amphibolite, then to biotite, and finally to vermiculite–biotite. The vermiculite–biotite is shallow and grades into biotite at depth. Whatever the source of the mafic layers that are now vermiculite, the occurrences of vermiculite in gneisses and schists have similar characteristics that are distinct from those of other types of vermiculite occurrences. Pegmatites and other sialic rocks are associated with these deposits but no zoning of the type found in the dunite and serpentine bodies is apparent.

CARBONATE OCCURRENCES

Two examples of this type of occurrence are to be found near Llano, Texas (Clabaugh and Barnes, 1959). Magnesite at the Texas Mines, south of Llano, contains vermiculite (which is almost the pure magnesian end member)

where the magnesite has been exposed to weathering. A few inches beneath the surface the magnesite contains unaltered phlogopite. Magnesite that was mined during World War II and left in stock piles near the pit now contains vermiculite that apparently has resulted from the action of rain water on the phlogopite. A marble that contains flakes of vermiculite is in a road cut along the road from Llano to Castell.

The present author has identified vermiculite from two localities north of New York City. A very light-colored vermiculite occurs in layers in the Wappinger marble at Torry Cave on U.S. Hwy. 7, just south of Gaylordsville, Connecticut. Within the carbonate, which is mostly calcite, the vermiculite is associated with phlogopite and talc. At Hawthorne Circle near Hawthorne, New York, vermiculite occurs in the Inwood marble where it is exposed in a road cut. At both of these occurrences the vermiculite as well as the other silicates appear to have been derived from silty layers in the original limestone at the time of regional metamorphism.

Another occurrence of vermiculite in marble is reported in Uganda by Davies and Bisset (1947) and by Taylor (1955).

GRANITIC OCCURRENCES

Granite from Daggett Pass, southeast of Lake Tahoe, California, contains flakes of black micaceous material that puff when heated in a flame. X-ray diffractometer patterns of these flakes show a broad reflection at 8° 2θ, indicating that the material is probably a randomly stacked, mixed-layer vermiculite–biotite with more biotite than vermiculite. This probably is a widespread form of vermiculite that undoubtedly has been identified as biotite many times over.

ORIGIN OF VERMICULITE

A census of the literature on the geology of vermiculite occurrences shows that the investigators are rather evenly split three ways: those concluding that vermiculite formed by the action of hydrothermal solutions ("H" in the References), those believing vermiculite formed by the action of supergene solutions on biotite and phlogopite ("S" in the References), and finally those believing that vermiculite formed by hydrothermal or supergene solutions, or both ("HS" or "SH" in the References, depending on emphasis).

Hydrothermal

Association with high-temperature rocks and minerals.—Vermiculite is found in close association with pegmatites and with talc, hornblende, anthophyllite, tremolite, chlorite, kyanite, sillimanite, garnet, and other high-temperature minerals, thus suggesting that the vermiculite itself is a high-temperature mineral.

Pseudomorphs after high-temperature minerals.—Hagner (1944) reports the occurrence of vermiculite pseudomorphs after hornblende, garnet, and other

high-temperature minerals, once more suggesting that vermiculite itself is a high-temperature mineral.

Vermiculite at depths in excess of 200 ft.—Although vermiculite can be shown to extend to shallow depths in some localities, it extends to considerable depths in others. At Libby, Montana, the grade of the ore 200 ft below the surface is as good as it is at the surface.

Supergene

Room temperature alteration of biotite to vermiculite.—It has been reported that biotite partially alters to vermiculite when placed in a 0.001 M solution of $MgCl_2$ at room temperature for 56 hr (Bassett, 1959).

Formation of chlorite from vermiculite under simulated hydrothermal conditions.—Roy and Romo (1957) placed vermiculite in a hydrothermal bomb and exposed it to 10,000 psi pressure and 300° C temperature. Under these conditions the vermiculite partially altered to chlorite. Roy and Romo concluded that since vermiculite does not continue to exist under hydrothermal conditions it could not form under hydrothermal conditions. If more magnesium had been made available in the system, the vermiculite might have altered completely to chlorite (Bassett, 1959).

The inhibiting effect of potassium on the formation of vermiculite.—Bassett (1959) has shown that biotite will not alter to vermiculite in even a molar solution of $MgCl_2$ at 100° C if potassium is present in concentrations greater than 0.04 M. He concludes from this evidence that vermiculite could not have formed at the time of pegmatite or syenite intrusion, as potassium-rich solutions would emanate from these materials.

Shallow depth of many vermiculite deposits.—A change from vermiculite to biotite with increasing depth can be found at the Day Book, Bush, Texas Mines, Tigerville, and other occurrences. As mentioned earlier, however, the "vermiculite" at Libby, Montana, extends to a depth of 200 ft with no significant change in the vermiculite–biotite ratio.

Vermiculite resulting from 15 years of weathering.—Phlogopite-bearing magnesite left in stock piles at the Texas Mines since World War II is now vermiculite-bearing magnesite. This alteration from phlogopite to vermiculite clearly resulted from the action of supergene solutions.

Remnant masses of biotite in the pyroxenite bodies.—Remnant masses of pyroxenite in these bodies indicate that the alteration of the pyroxenite was incomplete. Remnant masses of biotite that are separated from the unaltered masses of pyroxenite indicate that the alteration of biotite to mixed layer biotite–vermiculite was incomplete. The spatial separation of these masses strongly suggests that the alteration of the biotite to mixed-layer biotite–vermiculite took place at a different time and by a process different from the alteration of the pyroxene to biotite (Bassett, 1959).

Association with kaolinite and deweylite.—Kulp and Brobst (1954) found that weathering apparently altered some of the feldspar in the pegmatite at the Day Book deposit to kaolinite. Bassett and Lapham (unpublished investigation) have identified deweylite intimately mixed with the vermiculite

at the Brinton Quarry, and Lapham (1961) considers deweylite a supergene mineral.

Vermiculite deposits rare or missing in glaciated regions.—Kulp and Brobst (1954) have pointed out that no vermiculite deposits of significant size have been reported in the ultramafic bodies of New England. They suggest that glaciation removed the New England vermiculite deposits. This could happen only if vermiculite deposits are relatively surficial features, i.e. the result of supergene solutions.

Proximity of vermiculite to pegmatite.—In an ultramafic body that has been intruded by a pegmatite dike, the vermiculite occurs in the zone closest to the pegmatite. If the vermiculite formed at the time of the invasion of the pegmatite, one would expect the vermiculite (a magnesium-, iron-rich mineral) to form in the zone close to the ultramafic and away from the pegmatite. The pegmatite at the time of intrusion must have produced alkali- and silica-rich solutions. As mentioned earlier, even low concentrations of potassium inhibit the formation of vermiculite and cause mica to form instead. The environment adjacent to a pegmatite is geochemically more favorable for the formation of mica than vermiculite at the time of the intrusion of the pegmatite. It seems logical then that the vermiculite which occurs in close proximity to a pegmatite intrusive did not form as a result of the pegmatite intrusion, but formed as an alteration of the mica at a later time.

The presence of chlorite in the vermiculite zone at the Brinton Quarry, Gregory Mine, and Carl Moss Mine suggests that chlorite as well as mica formed at the time of the pegmatite intrusion and that chlorite formed where magnesium was abundant, while mica formed where potassium was more abundant. An alternative interpretation might be that the chlorite and vermiculite formed simultaneously as a result of supergene alteration.

Vermiculite in the pegmatite.—At the Brinton Quarry streaks of biotite occur in the pegmatite. These streaks are small and were wholly contained within the pegmatite. Where streaks and pods of biotite were open to the contact between the pegmatite and the ultramafic, the biotite has been altered to mixed-layer biotite–vermiculite. All the original micaceous material in the pegmatite is believed to have been biotite at the time the pegmatite intruded. The biotite that was exposed to magnesium-rich supergene solutions from the serpentine was altered to vermiculite–biotite, while the biotite that was locked in the pegmatite was sealed from the magnesian supergene solutions.

The role of soluble magnesium and calcium salts.—It is worthy of note that the vermiculite found in close association with magnesium and calcium salts, as in the carbonate-type deposits, is 14 Å material unmixed with biotite or phlogopite. The vermiculite found in pyroxenites and amphibolites, on the other hand, apparently is not associated with soluble magnesium or calcium salts at the present time, and is in mixed-layer association with biotite and phlogopite. It is conceivable that magnesium and calcium salts were present originally in the pyroxenite and amphibolite deposits, but were dissolved by

supergene water that carried the magnesium and calcium ions to the biotite or phlogopite, altering it to mixed-layer vermiculite–biotite or vermiculite–phlogopite. Had more soluble magnesium and calcium been available, alteration of mica to vermiculite might have been more complete, as in the magnesite and dolomite deposits, but apparently the source of soluble magnesium and calcium was limited and the alteration was only partial, resulting in mixed-layer material. That the mixed-layer material alters readily to pure vermiculite when it is placed in a magnesium or calcium solution is easily demonstrated and lends strength to this argument.

Hydrothermal versus Supergene

Three major arguments favoring the hydrothermal origin of vermiculite have been put forward: association of vermiculite with high-temperature rocks and minerals, pseudomorphs of vermiculite after high-temperature minerals, and vermiculite occurrences at depths in excess of 200 ft.

That vermiculite occurs in association with high-temperature rocks and minerals is undeniable. However, it does not follow that the vermiculite also formed as a high-temperature mineral. Vermiculite can form just as easily as a low-temperature alteration of mica that formed as a high-temperature mineral. Essentially the same argument can be put forward for the occurrences of vermiculite pseudomorphous after high-temperature minerals.

Percolation of supergene solutions to a depth of 200 ft at Libby, Montana, is not unreasonable in view of the strong relief of the region. The mine at Libby is approximately 2000 ft above the Kootenai River, 3 miles away. The pyroxene, biotite, asbestos, syenite complex at Libby is probably much more permeable than most ultramafic bodies.

CONCLUSIONS

Investigations of field and laboratory evidence and a search of the literature have produced several convincing arguments favoring an origin of vermiculite by supergene alteration of biotite and phlogopite. Very few observations favoring a hydrothermal origin have been presented, and the ones that have been presented are just as easily explained by supergene alteration of biotite and phlogopite. The two sets of criteria are listed in Table 1.

There can be little doubt that supergene solutions are responsible for the vermiculite at some localities, especially those in which the quantity of vermiculite is clearly depth dependent or at which the formation of vermiculite has been observed over a period of years. At other localities the evidence is less certain, and while the evidence for the supergene origin of vermiculite may be more convincing, a hydrothermal origin cannot be ruled out altogether. It is not yet possible to make a generalization about the origin of all macroscopic-type vermiculite deposits, but it is the considered opinion of this author that all macroscopic vermiculites owe their origin to supergene alteration of biotite and phlogopite.

TABLE 1.—POSSIBLE CRITERIA FOR ORIGIN OF VERMICULITE

Hydrothermal origin
Association with high-temperature rocks and minerals
Pseudomorphs after high-temperature minerals
Extent of vermiculite to considerable depths (200 ft)
Supergene alteration of biotite and phlogopite
Room temperature alteration of biotite
Alteration to chlorite under hydrothermal conditions
Inhibiting effect of potassium on the formation of vermiculite
Shallow depth of many vermiculite deposits
Vermiculite resulting from 15 years of weathering
Remnant masses of biotite and pyroxene in pyroxenites
Association with kaolinite and deweylite
Vermiculite deposits rare or missing in glacial regions
Proximity of vermiculite to pegmatites
Vermiculite in pegmatites cutting ultramafics

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H = Hydrothermal
S = Supergene
HS = Hydrothermal and/or Supergene

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