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Reconnaissance Study of the Geology of U.S. Vermiculite Deposits— Are Asbestos Minerals Common Constituents?

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Abstract

Unusually high incidences of asbestos-related mortality and respiratory disease in the small town of Libby, Montana, have been linked to amphibole mineral fibers intergrown with the vermiculite deposits mined and milled near the town from 1923 to 1990. A study conducted by the U.S. Agency for Toxic Substances and Disease Registry concluded that mortality due to asbestosis in Libby mine and mill workers and residents during 1979 to 1998 was much higher than expected for a similar Montana or United States population group. Recent medical testing of past and present mineworkers and residents of Libby showed lung abnormalities in nearly one-fifth of the adult study participants. The U.S. Environmental Protection Agency, under Superfund authority, is completing sampling and cleanup of asbestos-bearing materials in the mine, mill, and town sites. The U.S. Geological Survey is conducting a study, reviewed herein, to investigate the mineral content of other U.S. vermiculite deposits and to determine if the amphibole asbestos minerals like those found in the Libby deposits are common in other vermiculite deposits.

Introduction

Vermiculite is a general term applied to a group of platy minerals that form from the weathering of micas by ground water. Their distinctive characteristic is a prominent accordion-like unfolding and expansion when heated to between 800° and 1,100°C, depending on the composition and content of the vermiculite-bearing material. After processing (heat expansion), the vermiculite material is very lightweight and possesses fire- and sound-insulating properties. It is thus well suited for many commercial applications. The Libby (also called "Rainy Creek" or "Zonolite") mine was the world's largest producer of vermiculite during its operation.

Several varieties of the amphibole mineral group occur within the vermiculite ore deposit that was mined near Libby. U.S. Occupational Safety and Health Administration (OSHA) asbestos standards include the regulation of five amphibole varieties, when these particular mineral compositions occur as particles that fit the regulatory definition of "asbestos fiber" (slender mineral particles with a specific length:diameter ratio). Two of the regulated "amphibole asbestos" minerals—tremolite and actinolite—are common as mineral fibers in the Libby mine rocks, along with fibrous particles composed of three other members of the amphibole group (winchite, richterite, and ferro-edenite).

The U.S. Geological Survey (USGS) has recently studied the composition of 101 vermiculite-rich, archived samples collected from 62 vermiculite mines and deposits in 10 U.S. States. The samples were collected as part of a survey of the Nation's vermiculite resources in 1947, 1966-1967, and 1975-1976. At each mine or deposit that was visited, a limited number of hand samples were collected to represent the crude vermiculite material ("ore"). Thus, these samples likely do not fully represent all the materials present at a given site. In the current study, portions of each sample were examined by X-ray diffraction (XRD), scanning electron microscopy (SEM), energy-dispersive spectroscopy (EDS), and electron probe microanalysis. Despite the reconnaissance nature of the original sampling, the mineralogical characterization of the vermiculite samples has so far yielded consistent results. These preliminary results indicate that amphibole fibers (consistent with "amphibole asbestos" as defined by OSHA) are not common in some types of vermiculite deposits, but the amphibole asbestos mineralogy similar to that found in the Libby deposit is not unique. Initial results suggest that vermiculite deposits that formed within geologic settings similar to the Libby deposit—relatively quartz deficient, potassium-, sodium- and calcium-rich igneous intrusions, typically zoned—may contain amphibole fibers with chemical compositions similar to that of the Libby deposit minerals. Also, vermiculite deposits found where masses of ultramafic rocks are cut by granite and (or) pegmatite can contain amphibole fibers. These relationships may help guide priorities for sampling, monitoring, permitting, and reclamation of active and historic vermiculite mines.

The Vermiculite-Asbestos Issue

The association of amphibole asbestos with mined vermiculite has received significant nationwide attention since November 1999, when the Seattle Post-Intelligencer newspaper reported that unusually high incidences of asbestos-related

Figure 1. Location of the Libby (Rainy Creek, Zonolite) mine near Libby, Mont., site of open-pit mining of vermiculite from 1923 to 1990.

mortality and respiratory disease occur in the small, vermiculite-mining town of Libby, Mont. (Schneider, 1999). Asbestosis and other respiratory diseases in Libby residents are thought to be directly linked to amphibole-asbestos minerals found in the vermiculite ore deposit, which was mined about 6 miles northeast of the town (fig. 1) from 1923 until mine closure in 1990 (106th Congress, 2000). The U.S. Agency for Toxic Substances and Disease Registry (ATSDR) reviewed death certificate data for Libby mineworkers and residents for the 20-year period of 1979 to 1998 (Dearwent and others, 2000). Their report concluded that "mortality in Libby resulting from asbestosis was approximately 40 to 60 times higher than expected" compared "with mortality statistics for the state of Montana and the U.S. population" (Dearwent and others, 2000). The ATSDR conducted medical tests of the mineworkers and past and present residents of Libby. Included were sets of chest X-rays, pulmonary function lung tests, and face-to-face interviews with each person to review his or her medical history. Results released in August 2001 reportedly showed lung abnormalities (such as pleural thickening or scarring of the lungs) in about 18 percent (994) of the 5,590 adults participating in the testing (McLaughlin, 2001; Smith, 2001). The U.S. Environmental Protection Agency (EPA), under Superfund authority, is sampling and performing remediation in the town of Libby, the mines and mill sites, and the surrounding area.

The Libby (also known as the "Rainy Creek" or "Zonolite") vermiculite mine was an open-pit operation that began as a modest producer in 1923 but grew to be a major part of the world vermiculite market. The Libby mine is estimated to have supplied more than 50 percent of the world's vermiculite output based on available U.S. and world production data between 1924 and 1990 (DiFrancesco and Potter, 2001). It processed an estimated 20,000 tons (18,000 metric tons (t)) in 1940, 150,000 tons (136,000 t) in 1950, and 200,000 tons (180,000 t) in 1970 (McDonald and others, 1986).

Regulatory Definitions of Amphibole Asbestos

The term "asbestos" is a commercial term that refers to a group of silicate minerals that will easily separate into strands of thin, strong fibers that are flexible, heat resistant, and chemically inert, and thus well suited for applications such as heat insulation (Cossette, 1984; Ross, 1981; Ross and others, 1984; Skinner and others, 1988; Zoltai, 1981). In a mineralogical sense, the two groups of asbestos minerals are (1) serpentines, with the only asbestiform variety called chrysotile, and (2) specific amphiboles, including crocidolite (riebeckite asbestos), cummingtonite-grunerite asbestos (commercially called amosite), and the asbestiform varieties of tremolite, anthophyllite, and actinolite. According to OSHA asbestos regulation standards, in order to determine the "asbestos" content of a particular mineral assemblage, an analyst should count mineral particles that

1. Are 5 µm (micrometers) or longer in length and display asbestos growth habit (length:diameter aspect ratios of at least 3:1, usually exceeding 100:1);

2. Lack longitudinal striations that suggest the particle is a "cleavage fragment" (Campbell and others, 1979; Zoltai, 1981), which are exempt from regulation based on a 1992 OSHA ruling (Occupational Safety and Health Administration, 1992, p. 24320); and

3. Are composed of one of the six regulated asbestos "mineral" phases—chrysotile, crocidolite (riebeckite), cummingtonite-grunerite ("amosite"), tremolite, anthophyllite, and actinolite. These mineral compositions and morphologies were encountered in occupational exposures associated with asbestosis.

The OSHA asbestos regulation also notes: "Asbestos fibers exist in bundles that are easily parted, show longitudinal fine structure and may be tufted at the ends showing 'bundle of sticks' morphology" (Title 29, Code of Federal Regulations, 1999 (29 CFR 1910.1001)). Another guideline, although apparently unofficial in its application, is that asbestos fibers are flexible, typically indicated by long, thin fibers that appear to bend but not break. True asbestos is thought to be extremely flexible and not brittle (Zoltai, 1981).

For this study, we applied the terms "fiber" or "fibrous" in accordance with OSHA asbestos regulation standards, which define asbestos fiber as: "A particle that is 5 µm or longer, with a length-to-width ratio of 3 to 1 or longer" (Title 29, Code of Federal Regulations, 1999 (29 CFR 1910.1001)). We further considered "fibers" to include only those mineral particles with diameters visually estimated to be 3 µm or less, because regulatory agencies worldwide have generally agreed that at about 3 µm or less the mineral fibers are most hazardous to the human respiratory system. (See Cossette, 1984, p. 34–36.)

Study Goals

Careful analyses and characterization of the Libby asbestos minerals will (1) complement toxicological studies that examine the effects of these minerals on the human respiratory system, and (2) assist regulatory agencies in the identification of potentially dangerous asbestiform amphiboles relative to less hazardous amphiboles. For example, the U.S. OSHA and the U.S. EPA asbestos regulations (Title 40, Code of Federal Regulations, Part 61 and Part 763; Title 29, Code of Federal Regulations, Part 1910 and Part 1926) do not specify asbestiform richterite and asbestiform winchite, which are common constituents of the Libby deposits, as regulated asbestos minerals. (See Wylie and Verkouteren, 2000; Verkouteren and Wylie, 2000.)

A study is underway by the USGS to analyze the mineralogy of vermiculite-rich samples collected from 62 U.S. vermiculite mines and deposits. The study is initially being conducted by reconnaissance sampling and analyses using XRD, SEM, EDS, and electron probe microanalysis. The purpose of the study is to determine how common the amphibole asbestos minerals, like those found at Libby, are in other vermiculite deposits, and if they occur in similar morphologies and compositions.

The Geology and Uses of Vermiculite

Vermiculite Geology

Vermiculite is a general term applied to a group of platy, mica-like, hydrated silicate minerals with the general formula $(Mg,Fe,Al)_3(Al,Si)_4O_{10}(OH)_2{\bullet}4H_2O$. Most vermiculite group minerals are the products of aqueous alteration of micas, primarily the biotite subgroup (annite-phlogopite); they pseudomorph the platy morphology of the replaced mica. Typically, the biotite grains alter to hydrobiotite or chlorite, then to vermiculite (Bush, 1976). Vermiculites encompass a wide range of chemical compositions, vary in color from light yellow to green to brown to black, and generally have a bronze hue. Their distinctive characteristic is a prominent exfoliation when heated from 800° to 1,100°C. When heated, the vermiculite plates expand at right angles to the cleavage (accordion-like) as the contained water is converted to steam. This forms elongated, worm-like, light-weight particles that trap air. Individual particles can expand from 6 to as much as 30 times their original volume in the longest dimension (fig. 2).

Vermiculite forms from the low-temperature, weathering alteration of micaceous minerals in the zone of ground-water circulation (Bush, 1976). Thus, commercial vermiculite deposits require an igneous or metamorphic host rock that contains an abundance of large mica crystals, especially biotite or phlogopite, which have interacted with ground water and (or) surface waters. Biotite forms in a wide range of igneous and metamorphic environments, and phlogopite occurs in metamorphosed carbonate and ultramafic rocks. Bassett (1959) attributed the formation of biotite and asbestiform amphiboles in the Libby deposits to the alteration of augite (in pyroxenites) by high-temperature silica-rich solutions. Vermiculite formation clearly postdates the high-temperature rock-forming processes (Bassett, 1959; Bush, 1976).

Bush (1976) categorized vermiculite deposits into three broad groups, as follows: (1) deposits within large ultramafic intrusions, such as pyroxenite plutons, many of which are zoned and are cut by syenite or alkalic granite and by carbonatitic rock and pegmatite; (2) deposits associated with small to large ultramafic intrusions, such as dunite and unzoned pyroxenite and peridotite, cut by pegmatite and syenitic or granitic rocks; and (3) deposits in ultramafic metamorphic rocks. The Libby deposits are of the first deposit type.

Vermiculite Uses and Production

Processed (heat-expanded) vermiculite material is very light weight and has fire- and sound-insulating properties. Therefore, it is useful material for many commercial applications. Processed vermiculite is commonly used as a light-weight aggregate in concrete or plaster to make low-weight, sound-deadening, fire-resistant walls, boards, panels, and coatings. Other common applications are in horticulture and agriculture, as an additive to mulch, potting soils, and growing mixes, and as a carrier and extender for fertilizers, pesticides, and herbicides (Potter, 2001).

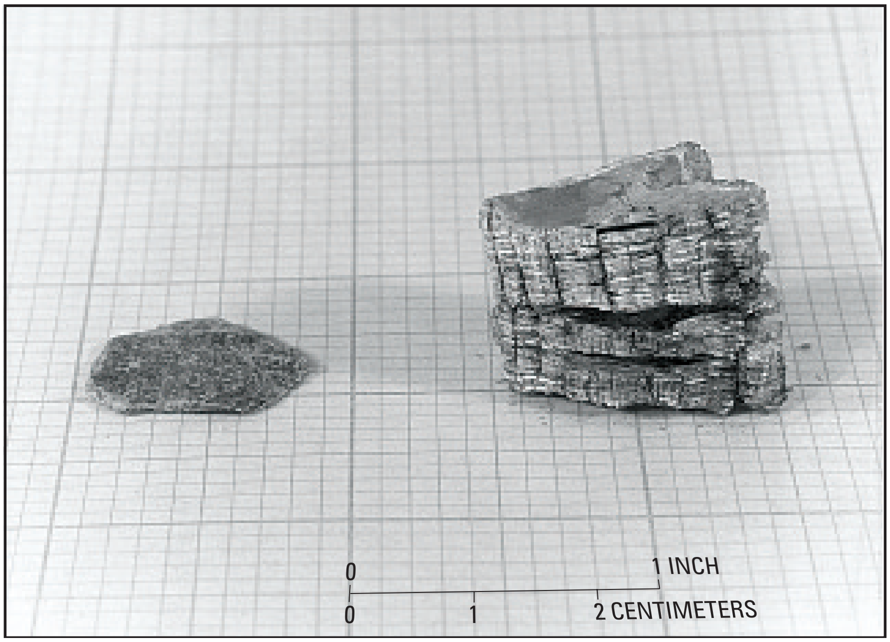

Figure 2. Examples of raw vermiculite (left) and processed (heat-expanded) material (right). When heated to 800°–1,100°C, the water naturally contained between the layers of the vermiculite structure is converted to steam. The steam pushes the crystal layers apart, expanding the vermiculite material from 6 to as much as 30 times its original thickness.

In 1999, South Africa led the world in production of vermiculite (218,000 t), followed by the United States (175,000 t), with much smaller production (each with 40,000 t or less) from People's Republic of China, Russia, Brazil, and several other countries (Potter, 2001). Three operations currently (2002) produce vermiculite in the United States—one operated by W.R. Grace & Co. near Enoree, S.C., one operated by Virginia Vermiculite Ltd. in Louisa County, Va., and one operated as a Virginia Vermiculite subsidiary near Woodruff, S.C.

Amphibole Mineralogy of the Libby Vermiculite Deposit

The Libby deposit contains amphiboles of several compositions that form intergrowths with the vermiculite and gangue rocks (fig. 3). Using current amphibole nomenclature, the amphibole compositions include winchite, richterite, tremolite, actinolite, ferro-edenite, and magnesio-arfvedsonite (this study; Wylie and Verkouteren, 2000). Winchite, richterite, tremolite, and actinolite represent subtle crystallographic and chemical compositional variations closely related to the tremolite-actinolite amphibole series (table 1), as explained by Wylie and Verkouteren (2000) and Verkouteren and Wylie (2000). Winchite, richterite, tremolite, actinolite, and ferro-edenite occur as blocky and elongate crystals, stubby and acicular cleavage fragments, and as fibrous (asbestiform) particles in the ores and gangue of the Libby vermiculite deposit (table 1; fig. 3). Magnesio-arfvedsonite, tentatively identified in samples from Libby, is found only as blocky crystals, and thus is not listed in table 1 as one of the fibrous amphibole minerals found in U.S vermiculite deposits. Fibrous winchite and richterite at other localities, as at Libby, are found associated with altered alkaline igneous rocks (Wylie and Verkouteren, 2000).

USGS Reconnaissance Study

The USGS is assembling a growing set of currently more than 100 samples of vermiculite. Most were collected from site visits to deposits throughout the U.S. by A.L. Bush in 1947, 1966-1967, and 1975-1976, and mostly during active mining. Additional samples were collected in 2001 at abandoned mine sites. The study samples are grab samples of vermiculite-bearing material ("ore") and are not fully representative of the materials mined or processed, or found in outcrop, at each particular deposit. Also, gangue and bedrock materials generally are not represented. Splits from 101 samples have been analyzed using SEM, EDS, and XRD methods. The samples analyzed are from deposits in Georgia (1 sample), South Carolina (27), North Carolina (14), Texas (21), Arizona (2), California (3), Wyoming (15), Idaho (4), Colorado (10), and Montana (4 non-Libby samples). The analytical results are compared to those for a representative suite of 30 samples that were collected from former mining operations at Libby.

Of the 101 vermiculite samples analyzed by SEM, 13 samples representing five vermiculite-rich districts were found to contain numerous fibrous amphibole particles. Two Colorado vermiculite districts—the Gem Park Complex, which straddles part of the Custer and Fremont County boundary, and the Powderhorn district in Gunnison County—contain an abundance of fibers semiquantitatively determined to be winchite, richterite, and riebeckite (fig. 4). A deposit in the Gold Hill district in Latah County, Idaho, contains abundant fibers of actinolite on the surface of vermiculite grains. Samples from the Smith mine vermiculite deposit in Converse County, Wyo., were found to contain fibrous tremolite and anthophyllite particles within a serpentine groundmass (fig. 5). The Addie district in Jackson County, N.C., contains abundant fibrous anthophyllite and tremolite associated with the vermiculite and talc. Amphibole fibers were found in minute amounts (a few fibers each) in 11 other samples from

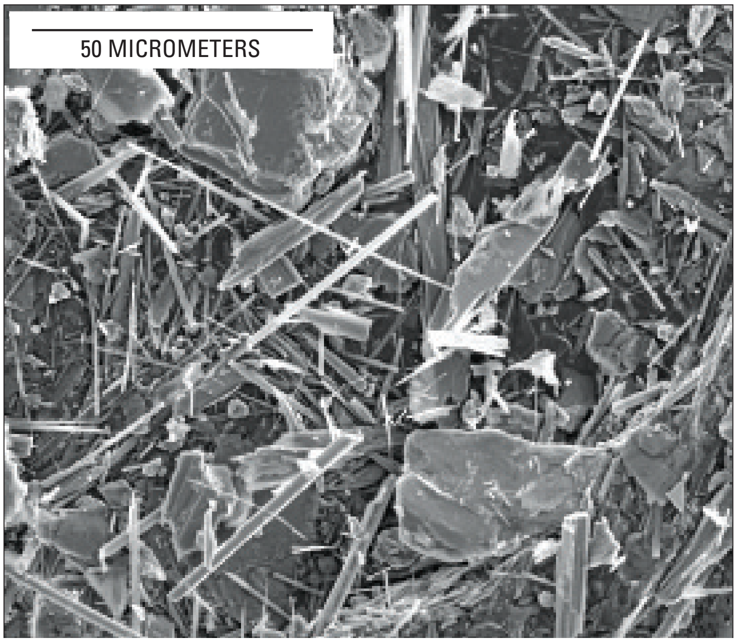

Figure 3. Scanning electron microscope (SEM) photograph of vermiculite ore material collected from an open pit of the Libby mine, Montana. The platy minerals have been identified as vermiculite, hydrobiotite, biotite, and phlogopite. Fibrous particles are amphibole phases with compositions identified as winchite, richterite, tremolite, actinolite, and ferro-edenite (table 1).

Table 1. Ideal compositions for the fibrous amphibole minerals found in U.S. vermiculite deposits.

[Cation ratios from Leake and others, 1997]

Mineral	Ideal cation ratios
winchite	$(Ca, Na)Mg_4(Al, Fe^{3+})Si_8O_{22}(OH)_2$
richterite	$Na(Ca, Na)Mg_5Si_8O_{22}(OH)_2$
tremolite	$Ca_2(Mg, Fe)_5\ Si_8O_{22}(OH)_2$ $Mg/(Mg+Fe^{2+}) = 1.0 – 0.9$
actinolite	$Ca_2(Mg, Fe)_5\ Si_8O_{22}(OH)_2$ $Mg/(Mg+Fe^{2+}) = 0.5 – 0.89$
ferro-edenite	$NaCa_2Fe^{2+}{}_5\ Si_7AlO_{22}(OH)_2$
riebeckite (crocidolite)	$Na_2(Mg, Fe^{2+})_3Fe^{3+}{}_2\ Si_8O_{22}(OH)_2$ $Mg/(Mg+Fe^{2+}) < 0.5$
anthophyllite	$(Mg, Fe^{2+})_7Si_8O_{22}(OH)_2$ $Mg/(Mg+Fe^{2+}) = 1.0 – 0.5$

nine deposits scattered throughout the U.S. The remaining vermiculite samples analyzed either lacked amphiboles or contained amphiboles that appear nonfibrous; these samples correspond to 48 deposits that generally fit the geologic characteristics of Bush's (1976) "type-2" or "type-3" deposits.

Thus far in the study, four of the six vermiculite districts that show abundant amphibole fibers associated with the vermiculite—the Libby vermiculite deposit, two Colorado areas, and one Idaho deposit—all have similar geologic settings. The Libby (Rainy Creek or Zonolite) deposit formed by alteration of a large zoned pyroxenite pluton having a central biotite-rich pyroxenite core. A younger mass of syenite cuts the outer zones of the pluton, and alkalic syenite dikes cut the biotitite core. A nearby small mass of nepheline syenite is present. Fenitization of the metasedimentary rocks surrounding the pluton suggests that a carbonatitic mass occurs at depth (Boettcher, 1967). One of the vermiculite-rich Colorado districts containing fibers is hosted by a zoned pluton—the Gem Park Complex—consisting mostly of pyroxenite and gabbro, cut by abundant carbonatite dikes and irregular masses with associated fenite (Parker and Sharp, 1970). The Gem Park Complex is cut further by minor dikes and bodies of lamprophyre, syenite porphyry, and nepheline syenite pegmatite. The complex is interpreted to contain a large carbonatite body at depth (Parker and Sharp, 1970; Armbrustmacher, 1984). The other Colorado vermiculite district with fibers is hosted by the Iron Hill stock (or "complex"), also described as a carbonatite intrusion. This stock is zoned, composed of mostly pyroxenite in the core. The stock's core is cut by nepheline-pyroxene garnet-rich rock. Sodic syenite and nepheline syenite intruded along the borders of the stock, and late dikes of nepheline gabbro and quartz gabbro cut the entire stock complex (Larsen, 1942; Temple and Grogan, 1965; Nash, 1972; Olson, 1974; Hedlund and Olson, 1975; Olson and Hedlund, 1981). The Idaho vermiculite deposit that contains actinolite fibers is hosted by a zoned hypabyssal stock—the Gold Hill stock. This stock has a core of mainly hornblende syenite with subordinate hornblende monzonite and hornblende syenodiorite (Tullis, 1944). Pyroxenite and syenite dikes are spatially associated with the stock and vermiculite deposit (Tullis, 1944).

The Smith mine vermiculite deposit in Wyoming contains tremolite and anthophyllite fibers formed at the contact zone between a granitic intrusion and massive serpentinite (Hagner, 1944). The amphibole fibers and biotite grains (which converted to vermiculite) apparently formed in a zone of metasomatic chemical exchange along the contact of the intruding granite with serpentine-rich country rocks. The North Carolina district—the Addie district—is associated with the Day

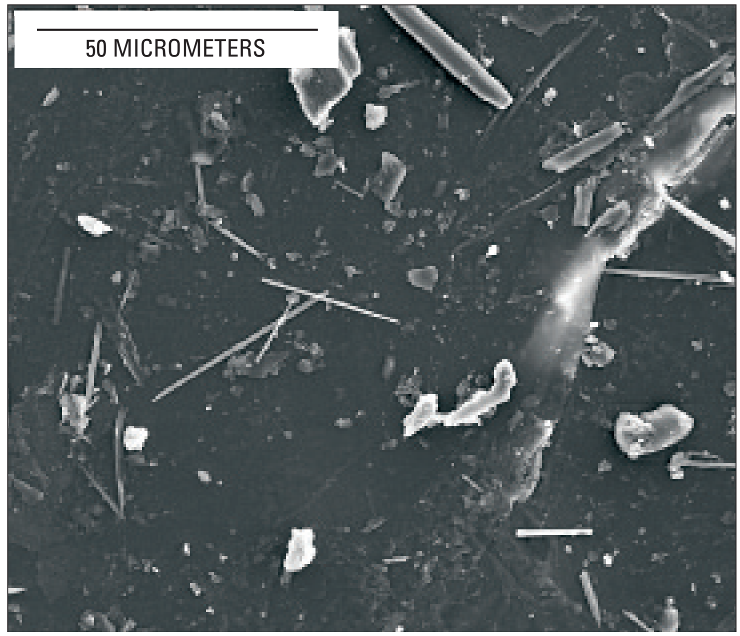

Figure 4. SEM photograph of vermiculite-rich material collected from Gem Park Complex, Colorado. Platy minerals visible are vermiculite and hydrobiotite. Fibrous particles are amphibole phases with compositions identified as winchite, richterite, and riebeckite (table 1).

Book dunite deposit and contains alteration zones composed of vermiculite (weathered phlogopite), fibrous tremolite and anthophyllite, and talc along serpentine-rich contacts between dunite masses and intruding pegmatites (Murdock and Hunter, 1946; Kulp and Brobst, 1954). The general geologic characteristics of these Wyoming and North Carolina vermiculite deposits correspond to Bush's (1976) "type-2" deposits. El Shazly and others (1975) described asbestos-vermiculite deposits in Egypt, and summarized similar deposits elsewhere, whose geologic characteristics are very similar to the Wyoming and North Carolina "type-2" vermiculite deposits.

Conclusions

The results of this mineralogical survey are preliminary, and the study sample set was collected in a reconnaissance fashion; therefore, additional, more representative sampling is planned. The results of this preliminary survey are consistent and suggest that fibrous amphiboles, in more than trace amounts, may not be common in the ore zones of some types of vermiculite deposits. However, the asbestiform amphibole mineralogy of the Libby deposit is not unique. The initial results suggest that vermiculite deposits associated with zoned, alkalic/calcic, quartz-poor plutons, especially those with characteristics of carbonatite intrusions, may be likely to contain fibrous amphiboles. Also, vermiculite deposits associated with ultramafic bodies cut by granite and (or) pegmatite deserve scrutiny, because contact metamorphic reactions along these contact zones often form serpentine-rich bodies that contain an abundance of amphibole fibers. These relationships may help guide priorities for sampling, reclamation, permitting, and monitoring of active and inactive vermiculite mines.

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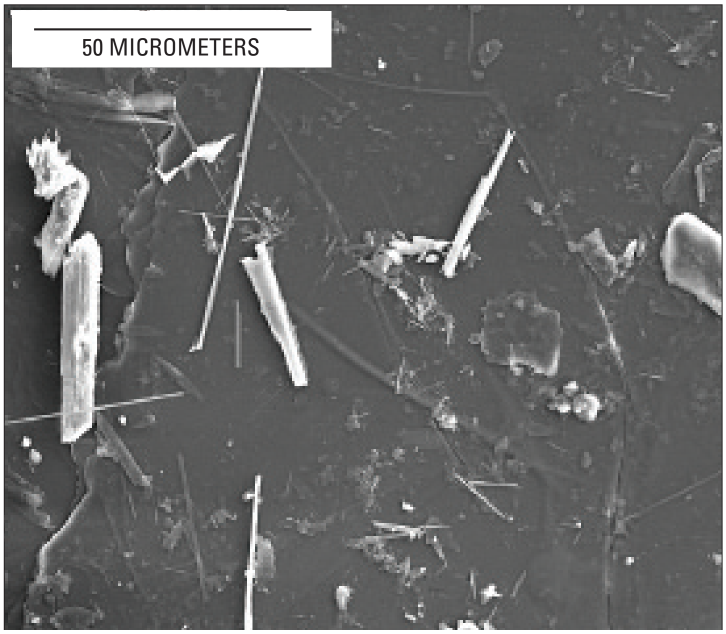

Figure 5. SEM photograph of a sample collected from Smith mine deposit, Wyoming. The fibrous particles have been identified as tremolite. Groundmass is mainly serpentine.

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