STEVENSITE AND TALC—HYDROTHERMAL ALTERATION PRODUCTS OF WOLLASTONITE

by

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ABSTRACT

Magnesium-bearing hydrothermal solutions moving along channelways created by faulting in the Willsboro, New York, wollastonite deposit have effected the conversion of wollastonite to layer-lattice silicates. Alteration products sheath faults in wollastonite and may be divided into three gradational but regular zones: (1) least altered (fresh) country rock composed of wollastonite with minor diopsidic clinopyroxene and grossularite-andradite garnet; (2) moderately altered rock rich in montmorillonite; and (3) strongly altered rock composed predominantly of talc.

X-ray diffraction, oscillating-heating X-ray diffraction, electron microscopy, infrared absorption, base exchange, and chemical analyses substantiate identification of the montmorillonitic alteration product, pseudomorphous after wollastonite, as stevensite. Stevensite, a trioctahedral magnesium montmorillonite, has been reported heretofore only in basaltic rocks where it has been formed by the action of magnesium-bearing solutions on pectolite. Pectolite and wollastonite have similar compositions and structures and may be similarly converted to stevensite in a magnesium-rich hydrothermal environment.

Talc, the dominant mineral in the most intensely altered zone bordering the solution channelway, was formed by the continuing action of magnesium-bearing solutions on stevensite. Stevensite has a defect structure, i.e., a deficiency in the total number of ions in octahedral coordination. The addition of magnesium from hydrothermal solutions to the octahedral layer of stevensite converts this mineral to talc, concomitantly freeing interlayer cations and water.

Stevensite is unstable and is converted to talc above a temperature of 275°C at water pressures of 690 bars. It is believed that hydrothermal alteration of wollastonite took place at temperatures near or below 275°C with stevensite forming in the cooler part of the reaction zone, and talc forming from stevensite at slightly higher temperatures nearer the solution channelway.

INTRODUCTION

During an intensive study of the mineralogy, petrology, and genesis of the Willsboro, New York, wollastonite deposit two areas, one in highly weathered surface rocks and the other in fresh underground exposures, were noted to be hydrothermally altered in the immediate vicinity of faults. Hydrothermal alteration of wollastonite-rich rocks has not been reported in geologic literature (to the writers' knowledge). This study was undertaken to determine the

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mineralogy of the hydrothermally altered wall rock, the physico-chemical environment of alteration, and to trace the course of alteration from fresh country rock to the most intensely altered zone.

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GEOLOGIC SETTING

The Willsboro wollastonite deposit is located two miles southwest of the town of Willsboro, Essex County, New York (Fig. 1). The wollastonite ore body is situated on the eastern flank of the Adirondack anorthosite massif and is encompassed in a northwest-trending belt of Precambrian Grenville Series sedimentary rocks (Fig. 1) that was engulfed, injected, partially assimilated, and metamorphosed by gabbroic anorthosite magma. Wollastonite-rich horizons are intercalated with mafic gneisses in this metasedimentary belt of rocks and one exceptionally rich unit is presently being mined.

The unaltered ore body is typified by a medium- to coarse-grained crystallloblastic aggregate of wollastonite–garnet–clinopyroxene with accessory apatite and sphene. White wollastonite, nearly pure CaSiO₃, constitutes approximately 64 per cent of the deposit. Nearly colorless to deep red garnet comprises about 21 per cent of the ore body and occurs as bands, lenses, and disseminated grains that are confined in composition to the grossularite–andradite series, Ca₃Al₂(SiO₄)₃–Ca₃Fe₂(SiO₄)₃. Green clinopyroxene of the diopside–hedenbergite series (variety salite) comprises about 15 per cent of the ore body and occurs in bands or lenses or as disseminated grains.

Faults and shear zones of uncertain, but probable Paleozoic age (Buddington and Whitecomb, 1941, pp.89–104), are ubiquitous in the country rock and can be recognized from a scale of microfaults in thin section to faults of major displacement in the country rock. The effects of faulting and subsequent movement of solutions along channelways created by faulting are, in general, confined to a brecciation of the country rock and to a slight retrograde alteration of wollastonite to aggregates of calcite and quartz. However, locally the action of hydrothermal fluids was more intense and gave rise to the unusual alteration products reported here.

WALL-ROCK ALTERATION

Physical and chemical changes effected by hydrothermal solutions passing through the Willsboro wollastonite deposit are restricted to one general environment, i.e., within or bordering faults and shear zones. Alteration halos, where developed, lie parallel to the walls of faults, but there appears
to be no direct relationship between the degree of shearing or brecciation and the intensity of alteration. Plate 1 shows a fresh underground exposure of altered wall rock along an east-west fault of minor displacement. The fault plane is reflected by a 1-inch vein of white calcite. Progressing outward and to
the left from calcite there is one-half an inch of greenish-brown, hard, siliceous fault gouge composed of quartz, calcite, and montmorillonite. Moving further outward the first true alteration halo is encountered. This halo is gray in color, two inches in width, and is composed predominantly of talc. A few fractures filled with calcite transect the halo. Hereinafter this alteration zone will be referred to as the inner, or talc, halo. Progressing further outward a light gray halo 1.5 in. in width is encountered. The color change between the inner and outer halos is not great but is sharp (Plate 1). The outer alteration halo is composed predominantly of montmorillonite and will be referred to as the outer, or montmorillonite, halo. The outer halo merges with relatively fresh wollastonite country rock.

The physical effects of the altering solutions are numerous. The color change is from white in the unaltered wollastonite to gray in the inner halo. A great change in grain size has occurred. The wollastonite country rock, ranging in texture from medium to very coarse grained, has been reduced to a very fine-grained aggregate of layer-lattice silicates (Plates 2 and 3). However, an increase in grain size may be noted for calcite as the fault is approached. The country rock has been softened as a result of the formation of the fine-grained aggregate of phyllosilicates and porosity and permeability have been changed as a result of the alteration. The outer montmorillonitic halo may be described as a clay inasmuch as it is a natural earthy, fine-grained material that develops plasticity when mixed with a limited amount of water.

**Fresh Country Rock**

Unaltered wollastonite-bearing rock of the ore body has been described above. The chemical analysis of unaltered wollastonite wall rock (Table 1) may be deemed typical of the bulk chemical composition of the ore body. The sample for analysis, however, was collected from an area immediately to the left of outer alteration halo, Plate 1.

**Outer Montmorillonite Halo**

The outer light gray halo is composed of montmorillonite, calcite, quartz, and talc with relics of garnet and salite. Wollastonite has been completely destroyed and is replaced by a very fine-grained aggregate of montmorillonite and quartz with minor calcite and talc (Plate 2). Talc is absent at the outer portion of the montmorillonite halo, but becomes more abundant at the expense of montmorillonite in samples closer to the channelway. Garnet appears fresh and shows only slight effects of brecciation. Salite is slightly altered to talc along cleavages and grain boundaries.

X-ray analyses of samples from the outer halo confirm the complete absence of wollastonite and the presence of montmorillonite, quartz, talc, and calcite. Diffraction maxima for garnet (Gr$_{76}$And$_{24}$) and salite are also present. Suspensions of material from the outer halo contain long acicular particles suggestive of wollastonite cleavage fragments. X-ray analysis of
such particles yields a mineral composition of montmorillonite, quartz, talc, and rarely, calcite. The fact that montmorillonite is pseudomorphous after wollastonite is also demonstrable from electron micrographs (Plate 4). Acicular cleavage fragments of wollastonite (Plate 4A) retain their external morphology when replaced by montmorillonite (Plate 4B).

X-ray powder patterns of montmorillonite demonstrate that the mineral is trioctahedral on the basis of the (060) peak position (1.523 Å). Oriented slides of the minus 2-micron fraction (Fig. 2) have a broad basal (001) montmorillonite reflection at 14.9–11.9 Å that sharpens and shifts to 16.98–17.10 Å after treatment with ethylene glycol. Heating to 450°C for one hour collapses the montmorillonite (001) peak to a 9.6 Å spacing. From oscillating-heating X-ray diffraction data, the montmorillonite loses most of its interlayer water at 42°C and all interlayer water has disappeared by 123°C. The loss of (OH) water is initiated at 605°C and is complete at 734°C.

The data obtained by X-ray analysis connote a trioctahedral montmorillonite. Wollastonite is incapable of supplying divalent cations compatible with the size requirements of the octahedral layer of montmorillonite minerals. The conversion of montmorillonite to talc, shown below, implies that sites in the octahedral layer are predominantly filled with magnesium ions. This

<table>
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<th>Unaltered Wollastonite</th>
<th>Montmorillonite Alteration Halo</th>
<th>Montmorillonite Alteration Halo, -2μ Fraction</th>
<th>Talc Alteration Halo</th>
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N. D. = Not Detected
* = Spectrochemical Analysis
Analyst: M. E. Coller, Dept. of Geology, Indiana University.
PLATE 1.—Photograph of alteration halos developed to left of fault in wollastonite-rich country rock. Pencil point is at contact between the inner talc halo and the outer montmorillonite halo.
Plate 2.—Photomicrograph of outer alteration halo. Wollastonite is replaced by a fine-grained aggregate of montmorillonite with minor calcite, quartz, and talc. Relict grains of garnet (high relief) and salite (with cleavage) remain. Plain light.

Plate 3.—Photomicrograph of inner alteration halo. Remnant grains of garnet and salite with new calcite are set in a fine-grained matrix of talc. Outlines of replaced wollastonite grains are visible. Plain light.
The assumption is substantiated by the trioctahedral nature of the montmorillonite and is further confirmed by a chemical analysis of material from the outer halo (Table 1). The low weight percentages of alumina (1.80) and ferric oxide (0.68) are almost entirely consumed in the formation of grossularite-andradite; therefore Al	extsuperscript{+++} and Fe	extsuperscript{+++} are not in the montmorillonite lattice. The chemical analysis of the minus 2-micron fraction (Table 1) reveals reduced percentages of alumina (0.28) and ferric oxide (0.48); again, this demonstrates that montmorillonite must be essentially free of these constituents. The relatively low weight percentage of ferrous oxide (1.90) (0.30 in the minus 2-micron fraction) is used up in the formation of salite clinopyroxene. Magnesium is the
only remaining element in abundance and of the right size to form the octahedral layer in montmorillonite. The association of talc and montmorillonite precludes calculation of a theoretical formula for montmorillonite, but the X-ray data and inferences drawn from the bulk chemical analysis indicate that the magnesium-rich montmorillonite is stevensite.

Stevensite is a hydrous magnesium silicate of the montmorillonite group that has been found in the United States only in the Watchung basalt of New Jersey. Caillère and Hénin (1956) reviewed the problem of nomenclature for the magnesium montmorillonites and suggested that saponite and stevensite should be regarded as the end members of magnesium-rich montmorillonites that have, respectively, abundant substitution in tetrahedral layers and very little substitution in tetrahedral layers. Faust and Murata (1953, p.982) indicated that the charge deficiency in the stevensite lattice originates through a deficiency in the total number of ions in positions of octahedral coordination. They therefore postulated that a defect lattice, involving a number of vacancies in random distribution, must exist in the octahedral (brucite) layer of stevensite. Faust et al. (1959, p.365) noted that the defect structure explains the low cation exchange capacity of stevensite (37.6 meq/100 g). They further noted that the concept of stevensite as a defect structure fits all experimental data, whereas Brindley's (1955, p.242) mixed-layer structure (interlayers of talc in saponite) fails to explain all the data.

It is noteworthy that all known specimens of stevensite are of hydrothermal origin, being derived from pectolite by the action of magnesium-bearing solutions. Pectolite, Ca$_2$NaH(SiO$_4$)$_2$, and wollastonite, CaSiO$_3$ or Ca$_3$(SiO$_3$)$_3$, are similar in chemical composition, have similar triclinic cells (Prewitt and Buerger, 1962, p.200), and thus might be expected to yield similar alteration products under the influence of magnesium-rich hydrothermal solutions.

Base exchange, infrared absorption, and quantitative chemical studies were used in conjunction with the data presented above to substantiate the identification of montmorillonite as stevensite. The base-exchange capacity of montmorillonite formed in the hydrothermal environment was determined because the base-exchange capacity of stevensite (37.6 meq/100 g) is considerably lower than that of "normal" montmorillonite (80–100 meq/100 g). The manganese-saturation method described by Bower and Truog (1940, pp.411-413) was chosen because it is accurate, relatively rapid and simple, and requires only small samples. Briefly, the procedure is as follows: a 1.0 g sample of dried material is saturated with 1N manganous chloride solution, the exchangeable manganese removed by leaching with 1N ammonium acetate, and an aliquot is taken. The manganese content is determined colorimetrically after oxidation of Mn$^{2+}$ to MnO$_4$$^-$. The reader is referred to the work of Bower and Truog for a detailed description of the analytical procedure.

The base-exchange capacity of material from the montmorillonitic halo was determined on a 1.00 g, polymineralic sample. Inasmuch as it was impossible to isolate the montmorillonite for base-exchange determinations
the abundance of montmorillonite was estimated from amplitudes of diffraction maxima. Great quantitative accuracy is not claimed for relative abundances but the accuracy is believed sufficient to separate stevensite from "normal" montmorillonites.

The 1.00 g sample of material from the montmorillonite alteration halo, estimated to contain 35–40 per cent montmorillonite, gave a base-exchange capacity corresponding to 14.73 meq/100 g. If montmorillonite constitutes 35–40 per cent of the sample this corresponds to a base-exchange capacity of 42.1–36.8 meq/100 g for the montmorillonite. The base-exchange capacity therefore agrees well with that of stevensite.

To verify the identity of the interlayer cation(s) a rather elementary test was devised. A 2.0 g sample from the outer montmorillonite halo was saturated with a solution of guanidine hydrochloride. Guanidine ions are planar and readily replace the interlayer cations of montmorillonite, but will not replace other ions. After repeated shaking over a 24-hr period the sample was filtered and the filtrate subjected to quantitative analysis. Theoretically, any inorganic cations in the filtrate were derived from exchange positions of montmorillonite. Quantitative analysis revealed calcium, magnesium, and potassium in a ratio of 66:8:1, respectively. Assuming the clay–mineral lattice to be saturated initially with respect to exchangeable cations the quantitative analysis was recalculated to yield an exchange capacity of 40–45 meq/100 g for the montmorillonite. This is in good agreement with the base-exchange capacity obtained by the Bower and Trought method.

The infrared absorption pattern of material from the montmorillonite alteration halo (Fig. 3) compares favorably with that given by Faust et al. (1959, p.354) for stevensite. R. Roy and V. Stubican have viewed the infrared absorption patterns and agree with the writers' identification of stevensite as the montmorillonitic alteration product of wollastonite.

**Inner Talc Halo**

Transition from the outer halo to the inner, or talc, halo is not as abrupt as the rather sharp color change would indicate. X-ray analysis of a series of samples from the outer portion of the montmorillonite halo to the inner portion of the talc halo reveals a consistent increase in the amount of talc with a concomitant decrease in montmorillonite. In the outer halo the talc is poorly crystalline as shown by relatively broad, asymmetrical (002) and (006) reflections. The reflections become sharp and symmetrical in samples nearer the fault, and are an indication of a higher degree of crystallinity. Diffraction maxima for calcite, garnet, salite, and quartz are also present on diffractometer patterns of unoriented powder samples of the inner halo. Quartz is very limited in abundance and montmorillonite is not present except in the outer margins of the halo. X-ray diffraction patterns (Fig. 2, D and E) and the infrared absorption pattern (Fig. 3) of the –2µ fraction of the inner alteration halo show talc to be the dominant constituent.

In thin section (Plate 3) rocks of the talc halo are seen to be composed of
grains of new calcite and remnant garnet and salite grains set in an extremely fine-grained aggregate of talc. Garnet appears to be stable in this rather intense zone of alteration, but salite shows slight alteration to talc and calcite.

Combining the oxides of the chemical analysis for material from the talc alteration halo (Table 1) into the minerals known to be present gives a weight composition of 62.3 per cent talc, 14.4 per cent salite, 12.8 per cent garnet, 7.2 per cent calcite, and 3.3 per cent quartz or other silicates. Alumina and ferric oxide were used up in the formation of garnet; ferrous oxide and an equivalent amount of magnesia were used in the formation of salite; all carbon dioxide was used to form calcite; and the remaining magnesia was used for the formation of talc. The percentages given above are in good agreement with mineral percentages estimated from X-ray and petrographic studies.

Electron micrographs of talc from this zone of intense alteration show acicular particles of talc (Plate 4C) that are morphologically indistinguishable from cleavage fragments of wollastonite (Plate 4A) and acicular particles of stevensite pseudomorphous after wollastonite (Plate 4B). Electron micrographs therefore suggest that not only is stevensite pseudomorphous after wollastonite, but also that talc is pseudomorphous after stevensite.
Fault Gouge

A thin (0.5 in.) greenish-brown band of quartz, montmorillonite, and calcite is present between the talc halo and the calcite of the fault plane. The montmorillonite of this band is exceedingly well crystallized. Unoriented powders yield diffraction maxima for montmorillonite that are sharp and symmetrical, a relatively rare characteristic for montmorillonite. The (060) peak position reveals that the mineral is trioctahedral. X-ray patterns of both oriented and unoriented materials indicate that the basal (001) spacing is constant at 15.02 Å, and is shifted to 17.0 Å after treatment with ethylene glycol (Fig. 2, F and G). Oscillating-heating X-ray diffraction studies show that interlayer water is lost gradually. Complete loss of interlayer water occurs by 150°C and the structure collapses to a 9.53 Å spacing.

The trioctahedral montmorillonite of the greenish-brown band is believed to be similar in both properties and origin to the stevensite of the outer halo except that it has a much better degree of crystallinity. The constant basal (001) spacing of 15.02 Å, the collapsed-structure spacing of 9.53 Å, and the complete loss of interlayer water by 150°C are in exact agreement with the data of Rowland et al. (1956, p.89) for their monionic calcium montmorillonite. The guanidine method outlined above gives calcium, magnesium, and potassium as the interlayer cations in a ratio of 72:6:1, respectively; and a base-exchange capacity of 57.4 meq/100 g. The Bower and Truog method gives a more accurate estimate of the base-exchange capacity as 52.0–46.8 meq/100 g. The infrared absorption pattern for fault gouge (Fig. 3) confirms the identification of this montmorillonite as stevensite and the electron micrograph of fault gouge (Plate 4D) shows the external morphology of the montmorillonite to be similar to that of stevensite from the montmorillonite alteration halo (Plate 4B).

A band of calcite within the fault gouge marks the fault plane and the channelway for hydrothermal solutions. The calcite is nearly pure CaCO₃. X-ray analysis, in conjunction with the graph of Goldsmith and Graf (1958, p.97) yields a composition of CaCO₃ with approximately two mole per cent MgCO₃.

DISCUSSION AND CONCLUSIONS

A hydrothermal origin is postulated for the development of alteration halos that border faults in wollastonite. The field relations, occurrence, and mineralogy of the alteration zones confirm that heated waters charged with carbon dioxide and magnesium moved along channelways created by faulting. The source of the hydrothermal solutions is unknown.

Additional information on the character of the hydrothermal solutions and the course of the alteration can be obtained from chemical analyses of unaltered and altered country rock (Table 1). The most striking variation shown in the analyses is that for magnesia. The unaltered country rock is very
low in magnesia (0.78 per cent). As the solution channel (fault) is approached the percentage of magnesia increases rapidly, reaching 19.6 per cent in the talc halo. A majority of the magnesia in the unaltered country rock is present in salite clinopyroxene. Salite is converted to talc and calcite in the altered areas, but even here the conversion is not complete. Salite cannot supply the necessary amount of magnesium for the talc-rich alteration products; therefore, the hydrothermal solutions must have supplied magnesium to the country rock.

The alteration products in wollastonite have a zonal distribution and appear to have developed in a single stage of hydrothermal activity. The mineralogical and chemical variations within the wall rock reflect continuously varying conditions of the physico-chemical environment outward from the hydrothermal channel. During the gradual outward penetration of magnesium-rich hydrothermal solutions wollastonite was unstable and broke down to free calcium. The metasilicate chains of wollastonite reorganized in the presence of magnesium and water to form stevensite. Calcium was removed by the solutions and deposited nearer to, or within, the main channelway. As the halo of alteration advanced outward from the fault the continuing action of magnesium-rich solutions converted stevensite to talc. Stevensite, with a talc-like structure, requires only the addition of magnesium to its octahedral layer to be converted to talc. The conversion of stevensite to talc would free the interlayer cations and water.

The presence of alteration halos along only one side of the fault is indicative of post-alteration fault movement. There is no reason to suspect that alteration halos would not develop symmetrically around the channelway inasmuch as similar rocks were present on both sides. Post-alteration fault movement also explains the development of the narrow greenish quartz–montmorillonite–calcite band on either side of the calcite fault lining. During these subsequent fault movements montmorillonite-rich gouge was moved along the fault plane from areas of previously formed montmorillonite alteration products. Remnant hydrothermal activity improved the crystallinity of the gouge montmorillonite. The development of the narrow greenish-brown bands against unaltered, as well as altered, wollastonite proves that the gouge developed after hydrothermal alteration was essentially complete. Subsequent fault movement is shown also by numerous small calcite veinings that penetrate the alteration halos.

Information concerning the chemical environment at the time of alteration has been given above. The resultant mineral assemblages also provide an indication of the physical environment at the time of alteration. At a temperature of 230°C and a water-vapor pressure of 5000 psi (345 bars) Roy and Roy (1955, p.154) were able to synthesize magnesium montmorillonite. They subsequently noted (1955, p.162) that at a water-vapor pressure of 10,000 psi (690 bars) magnesium montmorillonite is stable below 250°C, but is converted to talc at 275°C. Mumpton and Roy (1956, p.338) reported similar results. They noted that stevensite has a hydrothermal stability of
250°C and is converted to talc above this temperature. They determined that synthetic stevensite is stable to 275°C.

It is impossible to determine with accuracy the water pressures at the time of alteration. However, the range of water pressures given above is compatible with field relations and the geologic setting of the wollastonite deposit. If water pressure was equal to load pressure at the time of alteration the talc-montmorillonite halos were formed at depths between one and two kilometers. Temperatures of alteration must have been slightly above 250–275°C immediately bordering the hydrothermal solution channel, and must have decreased away from the channel. Temperatures in, or slightly below, the range of 250–275°C are indicated for the outer montmorillonite halo.

REFERENCES


