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UNITED STATES DEPARTMENT OF THE INTERIOR

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Steatitization of serpentinite bodies in north-central Vermont

by

Alfred H. Chidester

U. S. Geological Survey
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TEI-729. The action of heat and of superheated steam on the tuff of the Oak Spring formation, by G. W. Morey. 13 p., 4 tbls.

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STEATITIZATION OF SERPENTINITE BODIES IN NORTH-CENTRAL VERMONT

by

Alfred H. Chidester

ABSTRACT

The talc deposits of north-central Vermont are associated with bodies of ultramafic rock, chiefly serpentinite, that are enclosed by metamorphosed sedimentary and volcanic rocks of Cambrian and Ordovician age; these stratified rocks form an essentially homoclinal succession in the east limb of the Green Mountain anticlinorium. The rocks at the localities described here are in the greenschist facies, and show only slight variations in grade of metamorphism between localities.

The talc deposits consist typically of a core of serpentinite surrounded by successive shells of talc-carbonate rock and of steatite, next to which the country rock is altered for several inches to chlorite rock (blackwall). The steatite zone is rather uniformly about 3 feet thick, but the talc-carbonate rock and serpentinite zones vary considerably in thickness and are locally very irregular.

Serpentinization preceded steatitization, and probably was unrelated to it. Steatitization coincided with and was an integral part of the episode of regional metamorphism. Comparison of equal volumes of parent rocks and of rocks formed during steatitization (by means of a "modified standard cell") indicates that talc-carbonate rock was formed almost solely as a result of carbon dioxide metasomatism, with almost no gain or loss of other constituents except water; steatite and blackwall were formed essentially independently of the talc-carbonate rock, though at the same time, by metamorphic differentiation at the contacts between the serpentinite and the enclosing siliceous schist. The exchange of material in the blackwall-steatite reaction was more complex than in the alteration of serpentinite to talc-carbonate rock, but most of the material introduced (principally Si) came from the schist within a few feet of the serpentinite, and most of the material expelled from the serpentinite (principally Mg) reacted with the schist immediately adjacent to the serpentinite.

INTRODUCTION

The U. S. Geological Survey has for several years been conducting a study of the belt of ultramafic rocks in north-central Vermont. A detailed report on the ultramafic rocks and associated talc deposits at three representative localities (Barnes Hill, in Waterbury; Waterbury Mine, in Moretown; and Mad River, in Fayston) has recently been completed (Chidester, in press). The present paper is based upon that report, but is devoted almost exclusively to an analysis of the steatitization process. For details of geology beyond the bare background here presented, and for complete documentation, the reader is referred to will appear in the full report.

The writer is indebted to Professors Hans Ramberg and D. Jerome Fisher of the University of Chicago for encouragement and guidance in the preparation of the present report, and to his colleague Wallace M. Cady for critically reading the manuscript. It is with pleasure that the writer takes this opportunity to express his thanks.

Chemical analyses were made in the laboratories of the U. S. Geological Survey by Wilbur J. Blake, Robert N. Eccher, S. M. Berthold, E. A. Nygaard, E. I. Goldenthal, Leonard Shapiro, Paul W. Scott, Harry F. Phillips, Elizabeth L. Hufschmidt, and Hy Almond.

GEOLOGIC SETTING

The ultramafic rocks of Vermont, part of a great belt that extends from Alabama to Newfoundland, occur sporadically from Massachusetts to Canada near the central meridian of the state, and chiefly east of the axial ridge of the Green Mountains. The ultramafic rocks are emplaced in schists, gneisses, phyllites, and greenstones of Cambrian and Ordovician age near the western border of a broad belt of generally nonfossiliferous metamorphosed sedimentary and volcanic rocks that range in age from Cambrian to Devonian. West of the belt in which the ultramafic rocks occur, in the Champlain valley, is a succession of fossiliferous marbles, quartzites, slates, and phyllites of Cambrian and Ordovician age. Granitic rocks are principally east of the belt of ultramafic rocks, and show no spatial relation to the pattern of serpentinization or steatitization. Unmetamorphosed mafic dikes occur sparsely throughout the state.

The dominant structural features of the belt of ultramafic rocks is the Green Mountain anticlinorium, near the western border of the schists, gneisses, phyllites, and greenstones. Most of the ultramafic rocks are emplaced in steeply dipping strata that strike a little east of north in the east limb of the anticlinorium; a few in the northern part of the state are west of the anticlinorial axis.

GEOLOGY OF THE TALC DEPOSITS

General geology

The ultramafic rocks with which the present report is concerned (see fig. 1) are representative of those in a low-grade metamorphic setting that have been completely serpentinized and extensively steatitized; they embrace the extent of structural and petrographic variations within that type. The enclosing rocks consist of schist, greenstone, and minor quartzite, in which bedding and schistosity dip predominantly steeply to the east or are vertical; in a few places folds as much as 100 feet in wave length are evident. The ultramafic bodies are irregularly lenticular and range in size from a few feet wide and several tens of feet long to as much as 700 feet wide and perhaps as much as 6,500 feet long. Some are known to have a vertical extent of more than 600 feet. All, except the very small pods, contain cores (some of which are complex) of serpentinite. The cores are surrounded by shells of talc-carbonate rock as much as several tens of feet thick, which are in turn enveloped by a sheath of steatite [1] from a few inches to more than three feet thick. The talc-carbonate rock zone is commonly irregular; the steatite zone is generally regular and about 3 feet in thickness. Tabular veins a few inches thick of coarsely intergrown talc and dolomite cut the talc-carbonate rock and serpentinite in a few places. Immediately outside the

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[1] "Steatite" is used throughout this report to designate a rock composed essentially of talc, and no implication of suitability for manufacture of insulators is intended; indeed, none of the steatite of this report is suitable for industrial steatite.

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steatite zone the ultramafic bodies are surrounded by a thin shell of chlorite rock generally less than half a foot thick, commonly referred to as "blackwall"; where the country rock is albitic the blackwall is separated from the unaltered schist by an irregular zone a few inches thick that contains schist with very abundant porphyroblasts of albite, referred to as "albite porphyroblast rock." In a few places, where calcareous or dolomitic rock borders the ultramafic bodies, tremolite rock occupies the position of the blackwall and steatite is poorly developed. The original intrusive contact of the ultramafic body is inferred, on evidence that is developed in the section entitled "The mechanics of steatitization," to have been a few inches inside the outer margin of the steatite zone.

Structural features

Structural features include continuous schistosity, spaced schistosity, slip cleavage, fracture cleavage, steeply plunging east-trending folds, gently plunging north-trending folds, lineations, shear polyhedrons, and layering in the serpentinite. Continuous schistosity is local in occurrence; in it, all the micaceous minerals in a layer are uniformly parallel; it is probably the oldest secondary feature in the rocks. Spaced schistosity is the predominant schistosity; in it, micaceous layers are traversed by tiny shear zones a fraction of a millimeter apart. It is of complex origin; probably it is contemporaneous, in part, with the east-trending folds, which antedate the Green Mountain folding, but in many or most places it has been recurrently renewed in later orogenic episodes. Some of the steep lineation is also related to the early folds. The north-trending folds, spaced schistosity, and most crinkle lineations are related to the Green Mountain folding. Shear polyhedrons are believed to have formed by shearing and brecciation of the ultramafic rocks during tectonic transport of the crystalline mass. Layering in the serpentinite is possibly an inherited feature of the primary ultramafic rock.

Petrography

The schist in the localities under consideration varies considerably in composition, but consists essentially of quartz, sericite, and chlorite, and is commonly somewhat albitic. Graphite is commonly a conspicuous component, and biotite and garnet are locally present, in a few places as major constituents.

Greenstone ranges in mineral composition from actinolitic greenstone, composed essentially of actinolitic hornblende, albite, and epidote, to chloritic greenstone composed essentially of chlorite, albite, and epidote. The greenstone approaches the chemical composition of basalt.

Carbonate rock ranges in composition from magnesite marble to dolomite and calcite marble. It is invariably impure, and shows all gradations into schist and greenstone.

Blackwall chlorite rock consists essentially only of chlorite, but sphene and ilmenite are persistent accessories. The tremolite rock and the talcose carbonate rock of the blackwall zone are highly variable and sporadic in distribution; the former is associated with dolomite and calcite marble, the latter with magnesite marble. Albite porphyroblast rock contains features common to both schist and blackwall, but the dominant feature is the densely porphyroblastic character.

Serpentinite contains antigorite as the sole essential constituent, but magnetite and talc are common accessories, and chromite a relatively rare one. Veinlets of chrysotile asbestos are not uncommon, but are negligible in quantity.

Steatite is uniform in mineral composition, talc-carbonate rock is grossly uniform but variable on a small scale. Talc is the sole essential constituent of the steatite, talc and magnesite are the essential constituents of the talc-carbonate rock; magnetite is a common accessory of both rocks, chlorite common in steatite.

Selected chemical analyses of rocks and minerals associated with the talc deposits are given in table 1. Chemical compositions and modes of an idealized suite consisting of albitic schist, albite porphyroblast rock, blackwall, steatite, talc-carbonate rock, and serpentinite are given in parts A and B of table 2.

The compositions of minerals of significance to consideration of regional metamorphism and the steatitization process are as follows, as deduced from optical properties and chemical analyses:

The plagioclase in all the rocks is nearly pure albite, $NaAlSi_3O_8$.

Most of the epidote is pistacite, approximately $Ca_2Al_{2.4}Fe'''_{.6}Si_3O_{12}(OH)$, but tiny inclusions in albite appear generally to be clinozoisite.

Biotite varies widely in composition, depending upon rock type.

Actinolitic hornblende in the greenstone is uniformly near the composition $Na_{.2}Ca_{2.0}[Mg_{3.7}Fe''_{1.1}(Al,Fe''')_{.2}](Si_{7.6}Al_{.4})O_{22}(OH)_2$.

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Most of the chlorite in the schist and greenstone has a composition of about $(Mg_{2.1}Fe''_{2.6}Al_{1.3})(Si_{2.7}Al_{1.3})O_{10}(OH)_8$, but the Fe"/Mg ratio varies considerably and the Al content slightly. The chlorite in the blackwall is chiefly of the composition $(Mg_{3.5}Fe''_{1.2}Al_{1.3})(Si_{2.7}Al_{1.3})O_{10}(OH)_8$, but the Fe"/Mg ratio decreases sharply next to the steatite and increases next to the schist (see fig. 2).

Magnesite in the talc-carbonate rock and serpentinite ranges from about $(Mg_{.97}Fe''_{.03})CO_3$ to $(Mg_{.82}Fe''_{.18})CO_3$; the dolomite that occurs locally and in small amounts in the talc-carbonate rock has a composition of about $(Ca_{.50}Mg_{.45}Fe''_{.05})CO_3$. Rare rhombs of carbonate in the steatite are nearly pure calcite. Carbonate in the talc-carbonate veins is dolomite, with a composition, determined by chemical analysis, of about $(Ca_{.50}Mg_{.45}Fe''_{.04}Mn_{.01})CO_3$.

Talc in the steatite and talc-carbonate rock is uniform at each locality, but the ratio of Mg/Fe" appears to differ slightly among localities. An analyzed sample of steatite from the Waterbury Mine gave a calculated composition for the talc of $(Mg_{2.78}Fe''_{.25}Al_{.06})Si_{3.97}O_{10}(OH)_{2.16}$. Talc in the talc-carbonate veins consistently has a slightly higher ratio of Mg/Fe".

The composition of the antigorite, based upon chemical analyses of serpentinite, averages about $(Mg_{2.8}Fe''_{.15}Al_{.05})Si_2O_5(OH)_4$, but on the basis of indices appears to differ slightly between localities.

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Metamorphism

The mineral assemblages and the compositions of critical minerals indicate that the rocks at the three localities studied are in the greenschist facies of regional metamorphism, near the boundary with the epidote amphibolite facies. Retrograde metamorphic effects, chiefly the alteration of garnet to chlorite, are locally evident, but are negligible in the general consideration of the features of regional metamorphism. Slight differences in mineral assemblages, and the pattern of variation in composition of chlorite and biotite in critical associations, indicate that there are slight differences in grade between localities. The total range of variation in metamorphic intensity is small; the grade at the Barnes Hill locality is slightly below that at the Waterbury Mine, and the grade at the Mad River locality slightly below that at Barnes Hill. Figure 3 illustrates the pattern of variation for selected assemblages from the three localities.

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The inferred equilibrium relationships among the various mineral assemblages <u>permit</u> the conclusion that, at each locality, the steatite, talc-carbonate rock, serpentinite, and blackwall belong to the same metamorphic facies -- and therefore formed within the same broad range of P,T-conditions -- as the metamorphosed sedimentary and volcanic rocks; but the equilibrium relations, for the most part, do not <u>demand</u> that conclusion, as both talc and serpentine are stable far above the temperature of transition from the greenschist facies to the epidote-amphibolite facies. Though the geologic relations indicate that, at the time of emplacement, the serpentinite was not appreciably hotter than the enclosing rocks, the mode of origin subscribed to in this report for the serpentinite (discussed in the next section) places the origin of the serpentinite outside the realm of strictly regional metamorphism, and implies no direct relationship between serpentinization and the regional metamorphism of the rocks adjacent to the ultramafic bodies. Steatitization, on the other hand, appears, on the basis of all the available evidence, to belong to the episode of regional metamorphism. The evidence for this conclusion and the operation of the steatitization process, which constitutes the main theme of this paper, will be taken up in detail in the section entitled "Steatitization."

ORIGIN OF THE ULTRAMAFIC IGNEOUS ROCKS AND SERPENTINITE

The problem of the mode of origin of the ultramafic rocks has long been a perplexing one. A critical review of the problem is beyond the scope of this report, and a brief statement of leading current views will suffice as a basis for the succeeding detailed discussion of the steatitization process.

One school of petrologists has held that magmas of ultramafic composition are formed by partial refusion of a peridotite layer in the earth (see Hess, 1938; Buddington, 1943), and that peridotite and related ultramafic rocks, including serpentinite, may form by direct consolidation from such magmas. A second school, of which N. L. Bowen has been a leading spokesman (see Bowen and Tuttle, 1949), holds that ultramafic rocks are formed by fractionation of complex magmas of mafic composition. The second school considers that the ultramafic rocks were intruded in a crystalline state at low temperatures. The first school envisages the rocks to have consolidated directly from a magma of their own composition; but it is possible that rocks so formed may have been mobilized and reintruded at relatively low temperatures.

The field evidence indicates that the ultramafic rocks under consideration were intruded at temperatures comparable to those of the enclosing rocks. The experimental data (Bowen and Tuttle, 1949) indicate that magmas of the composition of dunite and peridotite cannot exist at temperatures below about 1,000° C. The present status of the problem, then, appears to be that the ultramafic rocks were emplaced at their present sites at low temperatures and in the crystalline state. Regional geologic relations in Vermont suggest that the ultramafic rocks may have formed by fractionation of complex magma from which associated volcanic rocks of the region were derived; but they may, perhaps equally plausibly, be inferred to have been derived from a peridotite shell of the earth.

Serpentinization is believed to have occurred chiefly during tectonic transport and emplacement of the ultramafic rocks. The alteration to serpentinite would be a necessary consequence of intrusion into wet rocks under proper conditions of temperature and pressure, and would have been facilitated by shearing and granulation during transport. Conversely, serpentinization would have facilitated movement of the mass of ultramafic rock. Appreciable, but probably minor, serpentinization may have occurred prior to the time that tectonic transport was a significant factor; and very minor serpentinization of relict olivine may have occurred during steatitization (see below), appreciably later than the main serpentinization.

STEATITIZATION

Among modern students of the petrology of ultramafic rocks, Gillson (1927), Hess (1933a; 1933b), Read (1934), Bain (1934; 1936), Du Rietz (1935), Phillips and Hess (1936), and Wiik (1953) have made important contributions to the study of the steatitization process. These investigators agree generally on the broad features of the process, but disagree with one or another in varying degree concerning such details as the precise age relations of steatitization; geometric and genetic relations between the several rock types formed during steatitization, between them and structural and metamorphic features, and between them and their parent rocks; the source, nature, and role of the steatitizing solutions; the thermal history of the steatitization process; and the precise nature of the chemical changes accompanying steatitization.

Critical features and relationships

A critical review of the problem of the origin of talc deposits associated with ultramafic rocks is beyond the scope of this report. For present purposes it will suffice to summarize the critical features and relationships displayed by the deposits in north-central Vermont which are pertinent to the problem, and to attempt to reconcile them, where necessary, with opposing views.

Age of steatitization

Steatitization followed serpentinization of the ultramafic rocks and is unrelated to it. The geologic evidence clearly indicates the sequential relationship, but the separateness of the two processes, though suggested by the weight of geologic evidence, rests largely upon the demands of the over-all integrated picture of the intrusion, serpentinization, and steatitization of the ultramafic rocks and the metamorphic and tectonic history of the area. The talc-bearing rocks are in all instances distributed peripherally to a core (in some instances complex) of serpentinite, and it is generally evident on the basis of textural and geologic relations that the talc rock has replaced the serpentinite; nowhere does serpentinite surround a central core of talc rock, and nowhere does serpentine appear to replace talc. These features clearly indicate that serpentinization preceded steatitization at any given point, but they do not require the two processes to have been unrelated, because it is readily conceivable that a single episode of alteration would proceed in two steps that would result in a zonal arrangement and sequential relationship of the products of the two steps. The inference of separateness for the two processes rests chiefly, insofar as geologic relations are concerned, upon the fact that the extent of serpentinization is unrelated to the width of the steatite and talc-carbonate rock zones, even in large bodies such as at the Mad River locality, whereas a single episode of combined serpentinization and steatitization would be expected to produce such a relation. The evidence for separateness is not compelling, but the concept of two unrelated processes seems to account most simply for the facts, and fits best with the present over-all interpretation of the geologic history of the ultramafic rocks.

Two possible minor exceptions to the above statements on the relative ages of talc and serpentine should be noted: (1) Small, isolated flakes of talc disseminated sparsely and rather uniformly throughout much of the serpentinite may be stable relicts (Barth, 1952, p. 327) formed prior to steatitization in the manner discussed by Bowen and Tuttle (1949, p. 456-457) during cooling of the peridotite. Such stable relicts of talc would be especially likely in an ultramafic body that contained a relatively large proportion of pyroxene. (2) It is not entirely impossible, or improbable, that some serpentine may have formed as a byproduct of steatitization. With the large quantities of water released by the alteration of serpentine to talc and talc-carbonate (see below, p. 33-50), it is very likely that any relict grains of olivine, or even compact masses of dunite, that escaped the first serpentinization may have been partly or completely altered to serpentine during steatitization.

Relations of steatite and talc-carbonate rock

The total body of evidence developed in the following sections leads clearly to the conclusion that the talc-carbonate rock and steatite are of the same age and of related mode of origin, although the geologic evidence alone is inconclusive. Earlier investigators failed to make a clear distinction between steatite and talc-carbonate rock, and consequently appear not to have considered that they may have formed by distinctly different processes, and may not, therefore, be necessarily of identical age. Indeed, the spatial relationship of the steatite as a shell surrounding the talc-carbonate rock suggests a sequential relation in which the steatite has replaced the talc-carbonate rock, although admitting of the possibility that both formed at essentially the same time. The talc-carbonate rock and steatite intergrade, and the textural relations of the minerals neither support nor oppose a replacement interpretation. The geologic evidence, therefore, permits a choice in interpretation of age relations, between essential simultaneity of talc-carbonate rock and steatite, or a younger age for the steatite.

Inasmuch as the geologic evidence is inconclusive on the precise age relations between the talc-carbonate rock and steatite, a conclusion must be based upon other considerations. In the several arguments developed in the following sections it will become evident that the chemical and mineralogical changes involved in the formation of the steatite and talc-carbonate rock are of such a character that they lead to a simple picture of essentially simultaneous origin for them, as two distinct phases of a single process.

Distinguishing between steatite and talc-carbonate rock also leads to slightly different conclusions about their distribution with respect to position in the ultramafic body. Hess, for instance (1933b, p. 640), shows the "talc" as being thick at the bottom of the lens and progressively thinner upward. This relation appears to be generally true for talc-carbonate rock, but not generally so for steatite. Consequently, the nature and role of solutions in steatitization, discussed in detail in a later section, is envisaged to be somewhat different from that pictured by Hess and others.

Relations between steatite and rocks of the blackwall zone

The blackwall is considered to be of the same age as and genetically related to steatitization, but that conclusion is not immediately evident. Several apparently equally probable alternative modes of origin suggest themselves, such as contact metamorphism or alteration in connection with serpentinization.

The blackwall grades rather sharply into steatite, on the one hand, and into schist on the other. Where an albite or porphyroblast rock occurs between blackwall and schist, it, too, is gradational into the rock on either side. Textural relations, pseudomorphs, and relict minerals furnish clear evidence of replacement in the chronological order schist ---> albite porphyroblast rock ---> blackwall ---> steatite, and imply a sequential chronological development.

Where the wall rock of the ultramafic body is calcareous or dolomitic, tremolite occurs at the outer contact of the steatite zone; the tremolite shows no textural evidence of disequilibrium with talc in most places, but not uncommonly crystals of tremolite are invaded and extensively replaced by talc. In a few places, where carbonate rock borders the steatite, tremolite crystals locally appear to be partially replaced by carbonate. Talc in magnesite rock replaces the carbonate and other minerals. The relations are not as clear-cut as for the blackwall chlorite schist, but tremolite rock appears to have formed by the reaction between calcitic or dolomitic carbonate rock and ultramafic rock; talcose carbonate rock appears to have formed in somewhat siliceous magnesite carbonate rock at the borders of ultramafic bodies.

The fact that the steatite nowhere cuts across the blackwall indicates that the blackwall is probably related to steatitization, rather than to some earlier process such as serpentinization. The chemical changes in the rocks of the alteration zone (see below, p. 45) show conclusively that interchange of constituents between the zones has been an important factor in their development. Consideration of all the evidence, therefore, leads to the conclusion that the rocks of the alteration zone were formed essentially simultaneously during steatitization; the replacement relations indicate not so much chronological sequence as widening of the reaction zones with time, accomplished by replacement of the next outer zone. This conception of simultaneity for the various rocks of the zone agrees with that of Read (1934, p. 537) rather than with Phillips and Hess (1936, p. 348). Further consideration of this and of the mechanism of replacement will be deferred until consideration of the mechanism of steatitization (p. 33).

The blackwall is of remarkably uniform thickness at the margins of the ultramafic bodies, regardless of position. This statement assertion differs markedly from Hess' conclusion contention (1933b, p. 640) that the blackwall is very thin at the keel of a lens of ultramafic rock, and very thick at the top. Perhaps these very different conclusions, both based upon direct observation, are explained as follows: Hess' picture of the configuration of the blackwall zone appears to be based largely upon relations observed at Roxbury (Hess, 1933b, figs. 3 and 4, p. 640-641; Phillips and Hess, 1936, p. 349-351). R. H. Jahns' detailed mapping (unpublished) of many of the Roxbury ultramafic bodies shows conclusively that many of the ultramafic bodies are emplaced in greenstone (chlorite-epidote-amphibole schists). In particular, the ultramafic body figured prominently by Hess (1933b, fig. 4) and Phillips and Hess (1936, fig. 8) is bordered on the west by a greenstone. This greenstone is altered to blackwall chlorite for a few inches from the contact with the ultramafic rock, as shown by Phillips and Hess. However, their "outer part of the zone of altered country rock" (labeled "chlor.-ep.-amph." in their fig. 8) is not a part of the blackwall, but is "unaltered" greenstone. Hess' conclusion is a logical one under the conditions of exposure at the quarry figured, and it is only upon more extensive mapping that the identification of his "outer blackwall zone" with the greenstone becomes evident. It is thus believed that, if Hess' outer blackwall zone be discounted, his views on the configuration of the blackwall zone are readily reconciled with those in the present paper.

Relation of steatitization to structure and regional metamorphism

Structural features of the blackwall and steatite indicate that steatitization took place after the peak of tectonic activity but before movement had ceased entirely. The local obliteration of earlier schistosity in blackwall and steatite, and the apparent preservation of relict schistosity in many places in these rocks show that the steatite and blackwall chlorite postdate the schistosity; the evidence for recurrent movement along schistosity planes and the local development of fracture cleavage in steatite and blackwall (see also Chidester, 1953) indicate that some deformation followed the formation of steatite and blackwall. Steatitization, therefore, bears about the same chronologic relation to orogeny as does regional metamorphism, and must be of about the same age.

The replacement relationship between the blackwall and the metamorphic minerals of the schist (such as, especially, the albite porphyroblasts) indicates that steatitization occurred during or after regional metamorphism had attained its peak. As with the rocks of the reaction zone, however, the evidence admits of the possibility that steatitization and regional metamorphism were essentially simultaneous. Though minerals of the outer blackwall zone are obviously later than the minerals (the albite porphyroblasts, for instance) that they replace, the blackwall is not necessarily younger than the regional metamorphism: where both isochemical reconstitution of the schist and metamorphic differentiation between the schist and serpentinite take place contemporaneously, the rocks of the reaction zone necessarily bear a replacement relation to both the schist and the serpentinite. It has been demonstrated earlier that the steatite and blackwall probably formed at the same conditions of metamorphic intensity as the schist. Thus the sum of the geologic evidence appears to indicate that steatitization took place concurrently with regional metamorphism. This conclusion is supported by inferences concerning the nature and source of the steatitizing solutions (p. 66).

Faults in the vicinity of bodies of ultramafic rock may have facilitated access of solutions (mainly CO_2) during steatitization, and may thus have been important factors in triggering and influencing the steatitization process. Sheared zones may have facilitated dissemination of solutions into the serpentinite; however, they did not exist as open channelways along which fluids migrated easily, as is attested by the generally uniform zonal distribution of steatite and talc-carbonate rock, though locally abnormal development of talc-carbonate rock, steatite, and blackwall in synclinal troughs and at other favorable structural controls indicates minor "channeling" of migrating fluids.

Volume relations in steatitization

It has long been accepted by some that replacement in metamorphism occurs on an approximately volume-for-volume basis (Barth, 1952, p. 313), but agreement is not universal. Ramberg (1952, p. 92-95), for example, concludes that the volume-for-volume principle does not always apply. Poldervaart (1953, p. 481-504) questions the general validity of the equal-volume principle, particularly under conditions of regional metamorphism, and proposes a system of calculations based on the assumption that the content of $(Si,Al)O_4$ tetrahedra in a rock remain constant.

In most places the geologic evidence on the volume relations of the products of steatitization is inconclusive. However, several types of evidence suggest volume-for-volume replacement for each of the rocks of the alteration zone (blackwall, steatite, and talc-carbonate rock), whereas none of the evidence appears to indicate perceptible change in volume during replacement.

In numerous places at the Waterbury Mine, partial to complete replacement of albite porphyroblasts by the chlorite of the blackwall can be observed in the outer part of the blackwall zone. Even where the crystal of albite is very deeply and irregularly embayed, there is no evidence -- such as fracturing of the albite grain around the embayment -- to indicate that replacement was accompanied by significant change in volume. The textural relations of the chlorite to other minerals of the schist are generally inconclusive, but there is no reason to suppose, and no evidence to suggest, that they differed essentially from those shown by the albite.

Relict layering is preserved in a few thin sections of blackwall. Comparison of the pattern of the relict layering with that of the adjacent schist indicates that no large change in volume has occurred, but generally admits the possibility of moderate change. The best place for accurate evaluation of evidence for or against change in volume is where contacts between the schist and blackwall and steatite cut across layering in the schist, and relict layering is preserved in the blackwall and steatite. At the Mad River locality, layering in the schist can be traced across a slightly crosscutting, fairly sharp contact with the blackwall for as much as an inch into the blackwall without evidence of discontinuity or systematic distortion of the layering. The best megascopic evidence of this sort is at the old East Granville talc mine (see Chidester, Billings, and Cady, 1951, p. 25), in northeastern Granville township, where one contact of a small lens of steatite cuts across layering in the schist at an angle of as much as 20 degrees. Relict layering in the steatite and blackwall can be traced clearly into the unaltered schist. None of the layers show discontinuity or systematic distortion at the boundaries of the different rocks. If any volume change had occurred during replacement, it is evident that, on either side of a given undistorted layer, successive layers should be displaced or distorted at the contact by increasingly large amounts in such a manner that the reference layer marks a plane of symmetry -- unless, of course, it can be assumed that all the expansion took place parallel to the layers. There appears to be no reason why such an assumption should hold; therefore, inasmuch as the layering exhibits no discontinuity or regular distortion pattern at contacts, it appears that replacement in the formation of blackwall and steatite was on a volume-for-volume basis.

Textural features around irregular embayments of steatite into chlorite rock and of talc-carbonate rock into serpentinite furnish further evidence for volume-for-volume replacement. Such irregular embayments of talc into chlorite are common at the scale of a thin section, particularly in inclusions of chlorite rock within steatite. A particularly instructive example of serpentinite embayed by talc-carbonate rock was pointed out by R. H. Jahns at one of the verde antique quarries at Roxbury. The embayment was very irregularly sacklike and more than 10 feet in greatest dimension. The "mouth" of the sack, at the border between the talc-carbonate rock zone and the serpentinite zone (the embayment is considered to lie within the general limits of the serpentinite zone) was perhaps a foot across -- considerably narrower than the average diameter of the embayment. Nowhere around the embayment of talc-carbonate rock was there evidence in the serpentinite of expansion or contraction during the alteration of serpentinite to talc-carbonate rock. Similarly, none of the steatite tongues and embayments shows evidence of change in volume during the alteration.

The positive evidence is thus seen to favor, in every case, constant-volume relations during formation of blackwall, steatite, and talc-carbonate rock.

Temperature relations during steatitization

Most earlier investigators have concluded that the geologic relations, particularly the textural relations among minerals considered to be diagnostic of different temperatures of formation, indicate marked changes in temperature during the course of steatitization. Bain favors a rising-temperature sequence, whereas Hess, Phillips and Hess, Burfoot, Du Rietz, and others favor a falling-temperature sequence. Read, however, concludes that the different zones were formed "simultaneously," and presumably, therefore, considers them to have formed at the same temperature conditions.

It has been shown in foregoing sections that the evidence from geological relations and mineral assemblages indicates that all the rocks formed during steatitization were formed under the same physical conditions as the adjoining schist, and that there was no significant temperature change during steatitization. These conclusions are reconciled with those of earlier investigators as follows:

It appears that both the rising-temperature and the falling-temperature hypotheses can be attributed to the idea that replacement of one mineral by another is almost invariably due to changes in temperature, particularly if one of the minerals is commonly regarded as indicative of higher temperature than the other -- as is the case for tremolite compared with talc, or biotite compared with chlorite. Experimental evidence shows, however, that some of the so-called low-temperature minerals, such as talc, are stable at relatively high temperatures (see, for instance, Bowen and Tuttle, 1949, p. 449), and cannot be taken unconditionally as indicating low temperatures (see Ramberg, 1952, p. 145).

Obviously, then, the sequence of alterations among minerals at the borders of the ultramafic bodies, such as of tremolite altered to talc, or chlorite to talc (and, incidentally, of biotite to chlorite in Phillips and Hess' (1936, p. 341) high-temperature type of deposit) may reflect only changing bulk composition -- or, more specifically, changing chemical potential of mobile constituents -- at a given point, rather than changes in temperature. This statement becomes evident upon examination of equilibrium diagrams of the various minerals (for instance, Ramberg, 1945, p. 55; 1952, p. 64). The metasomatic changes in the reaction zones at the borders of the ultramafic rocks (see the following section) clearly are of the kind to bring about the replacement relations exhibited. Some local reversals of general trends in alteration sequences among the minerals -- such as in the replacement of tremolite by carbonate -- may possibly result from local variations in the chemical potential of H_2O or CO_2 or both. It, therefore, appears to be in accord with all the evidence to consider that steatitization and related changes were accomplished at essentially a constant temperature, though some minor alterations may have occurred during the period of falling temperature.

The mechanism of steatitization

In the foregoing section the evidence has been shown to indicate that steatitization took place at essentially constant volume, contemporaneously with and at essentially the same temperatures as regional metamorphism, and without notable change in temperature during the steatitization process. It now remains to determine as precisely as possible the mineralogical and chemical changes that accompanied the steatitization process, and, having determined those changes, to reconstruct as nearly as possible the operation of the process and to discover the probable sources of introduced constituents, particularly H_2O, CO_2, and SiO_2.

It has long been recognized that direct comparison of chemical analyses given in terms of weight percent of oxides cannot result in an accurate description of chemical changes that took place during metamorphism, because such a comparison does not take proper account of material introduced or lost during the metamorphism. The more generally used modern methods compare the contents of equal volumes of rock, computed from chemical analyses and density measurements (see, for instance, Lovering, 1949; Poldervaart, 1953, p. 482-490). The contents are stated variously in terms of atoms per cc of rock or of chemical compositions calculated to represent 1 cc or 100 cc of rock. Barth (1945; 1948; 1949; 1952, p. 82-85) has proposed a "standard cell" of 160 oxygen atoms (which contains about 100 cations) as a basis for petrologic calculations, on the basis that comparing rock volumes of equal oxygen content is approximately equivalent to comparing equal volumes of rock. Poldervaart (1953, p. 481-504) has recently proposed a method of petrological calculations based upon the assumption that the $(Si,Al)O_4$ content of rocks remains approximately constant during metamorphism.

Barth's "standard cell" method is not suitable for present purposes because the rocks under consideration differ widely in cell size, inasmuch as they are characterized by minerals of widely different equivalent volumes. Poldervaart's method is not applicable in the present study because the $(Si,Al)O_4$ content of the rocks has changed appreciably during steatitization.

In this report the chemical changes effected during metamorphism will be determined by comparison of volumes of rock of equal and standard size, designated the <u>modified</u> <u>standard</u> <u>cell</u>. In brief, and in general terms, the modified standard cell is a standard volume of rock that, for an average rock, contains about 100 electropositive ions. The volume is taken as precisely $2{,}064.8 \times N^{-1}$ cc (or approximately $3{,}427.2 \times 10^{-24}$ cc) where N is Avogadro's number, 6.0247×10^{23}; this volume is represented by a cube about 15.1×10^{-8} cm on an edge. The method is basically similar to other methods that compare equal volumes of rock, cited above. It stems, however, from preliminary attempts to apply Barth's method of the standard cell, and retains certain desirable features of Barth's method. For most rocks, the modified standard cell -- like Barth's standard cell -- contains approximately 100 electropositive ions (cations); therefore, changes among such ions may be considered generally to be roughly in terms of percentages of their total in the parent rock. Barth's method of denoting the contents of his standard cell as a "rock formula" has also been imitated.

pages 36, 37, 38 are in the pocket in the back of this volume.

Changes in mineral composition are readily determined by comparison of volume percentages of minerals in the rocks -- whether of figures obtained by direct methods by means of microscopic analyses of thin sections, or of figures obtained by computing modes in volume percent from chemical analyses. Where it is desirable to compare mineral and chemical changes, as in the present study, it is more satisfactory in many ways to compute volume percent modes from the chemical analyses, rather than to use measured modes.

Table 2 and figures 4 and 5 assemble, in idealized form, the basic data of the steatitization process, to show the mineralogical and chemical changes that occurred during the alteration of serpentinite to talc-carbonate rock and steatite, and of schist to blackwall and steatite.

The calculated chemical analyses (CA-5, CA-6, CA-7, CA-8, CA-9, and CA-10) and derived data are based upon chemical analyses of four suites or partial suites of specimens across contacts of talc deposits, several chemical analyses of separate specimens, and modal analyses of more than a dozen additional suites or partial suites of rocks across the contacts, and information derived from industrial tests relating to the carbonate content of talc-carbonate rock. The thicknesses of the various zones (blackwall, steatite derived from schist, etc.) are based mainly upon observation, but have been adjusted very slightly so that the gain or loss of Mg in the combined blackwall and steatite zones is zero, as actual suites of analyses suggest is the case. For the detailed information upon which the calculated analyses are based, for tabulations of the actual calculations, and for a description of procedures followed, see Chidester (in press). 8

P. 39 follows

The data presented in table 2 and figures 4 and 5 are interpretative to the extent that they are idealized. The changes for major mineral and chemical constituents appear to be solidly grounded. The changes in some of the minor elements and of the less abundant and less common minerals are less firmly established, as will be discussed below.

It is evident from the data of table 2 and figures 4 and 5 that the talc-carbonate rock on the one hand, and the steatite and blackwall on the other, formed essentially independently of one another insofar as chemical interchange and introduction of material are concerned -- though both alterations undoubtedly took place concurrently. It is readily conceivable, therefore, that in some ultramafic bodies access of CO_2 was very limited or nil (locally, or entirely), that no talc-carbonate rock was formed, and that only the blackwall--steatite reaction operated effectively. It seems probable, however, that the talc-carbonate rock reaction was always accompanied by the blackwall--steatite reaction, which was not dependent upon introduction of material from outside the system and which was subject to the same general limitations of temperature and pressure as the talc-carbonate rock reaction. Possibly, also, the talc-carbonate rock reaction had a catalytic effect upon the blackwall--steatite reaction because of the release of abundant water from the serpentinite.

The relations at one locality (Mad River) appear to illustrate particularly well both the separateness of the talc-carbonate rock and the blackwall--steatite reactions and the possible catalytic effect of the talc-carbonate rock reaction upon the blackwall--steatite reaction. At this locality, the steatite and blackwall zones are thin and talc-carbonate rock absent from midway along the borders of the ultramafic body; the steatite zone is thicker at the ends of the body where talc-carbonate rock is abundant.

Table 2 and figures 4 and 5 contain the full account of quantitative gains and losses among the several rock types during steatitization. They are generally self-explanatory, but some of the data are erratic and require qualification, even though idealized. The remainder of this section will be devoted to discussion of the more important features of the table and figures, and to examination of the processes and reactions by which the changes illustrated in them were brought about.

The talc-carbonate rock reaction

The alteration of serpentinite to talc-carbonate rock may be represented approximately by the equation

$$\underset{\text{Serpentine}}{2(\mathrm{Mg,Fe''})_3\mathrm{Si_2O_5(OH)_4}} + \underset{\text{Magnetite}}{x(\mathrm{Fe'',Mg})\mathrm{Fe'''_2O_4}} + 3\mathrm{CO_2} \dashrightarrow$$

$$\underset{\text{Talc}}{(\mathrm{Mg,Fe''})_3\mathrm{Si_4O_{10}(OH)_2}} + \underset{\text{Magnesite}}{3(\mathrm{Mg,Fe''})\mathrm{CO_3}} + \underset{\text{Magnetite}}{y(\mathrm{Fe'',Mg})\mathrm{Fe'''_2O_4}} + \mathrm{H_2O},$$

where the Mg:Fe" ratio may differ for each mineral, and x and y vary in such a way that the total (Mg - Fe" - Fe"') is the same for both sides of the equation. The reaction involved chiefly the addition of CO_2 and the loss of H_2O. This conclusion is evident in table 1-D. Gains or losses in other constituents appear for the most part to have been less than about 1 percent of the bulk composition of the rocks and may have been essentially zero for nearly all constituents except CO_2 and H_2O. The uncertainty arises from the fact that many of the constituents (such as Fe", Fe"', and Al) present in small amounts are associated in appreciable proportion with minerals, such as magnetite and pyrite, whose distribution is spotty in both serpentinite and talc-carbonate rock, so that practical sampling procedures are inadequate for evaluating relative changes in those constituents. The case for each constituent will be discussed fully in the section on "Geochemistry of the rocks affected by steatitization," p. 53-65.

The calculations based upon the assumption of constant-volume replacement of serpentinite by talc-carbonate rock agree surprisingly well with field observations with respect to the proportion of talc and carbonate in talc-carbonate rock. In particular, the calculations agree strikingly with observed differences in carbonate content between Barnes Hill and Waterbury Mine (Chidester, in press). Such agreement appears to furnish strong support for volume-for-volume replacement, as does also the simplicity of the alteration process (in which the formation of talc-carbonate rock is regarded as being brought about simply by addition of CO_2 and loss of H_2O) that results from assuming constant-volume replacement. Such a conclusion would mean, if Si, Mg, and Fe were neither gained nor lost, that the composition of the talc and carbonate, and the proportion and composition of magnetite, along with the degree and nature of the alteration of the accessory minerals, varied in such a way that constant-volume relations were maintained. Thus, the volume relations would be the immediate determining factor (within limits) controlling the composition of the talc and carbonate, and the proportion of magnetite; but the composition of all would rest fundamentally upon the composition of the original serpentinite.

If maintenance of constant-volume relations in the alteration of serpentinite to talc-carbonate rock is of paramount importance, the reaction can be represented essentially according to the equation

$$aV(\text{serpentine}) + (1-a)V\,(\text{magnetite}) + \underset{\rightarrow}{CO_2} \dashrightarrow$$

$$bV(\text{talc}) + cV(\text{carbonate}) + (1-b-c)V(\text{magnetite}) + \underset{\rightarrow}{H_2O};$$

where a, b, and c are fractions that total less than 1 on either side of the equation, and V represents equal volumes of serpentinite and talc-carbonate rock; the short arrows under CO_2 and H_2O signify that they are mobile constituents that do not enter into considerations of volume relations. The volume proportion of talc in talc-carbonate rock depends upon the proportion of Si (and that probably negligible proportion of Al that may substitute for Si in talc) in the original rock, which controls the molecular proportion; upon the ratio of Mg:Fe" in the talc, which affects the equivalent molecular volume; and upon the proportion of accessory minerals. A marked sensitivity to pressure in the equilibrium relations between talc, carbonate, and magnetite is implied. Adjustment in the system would appear to depend chiefly upon variation of the Mg:Fe" ratio in the different minerals and upon changes in the Fe''':Fe" ratio (manifested by changes in content of magnetite) in the system. A necessary corollary to change in the Fe''':Fe" ratio is that O was mobile. A logical inference of the entire line of reasoning is that the chemical potential of O is significantly sensitive to pressure, and that this sensitivity is one of the more important factors in maintaining constant-volume relations in the talc-carbonate reaction. The variation of equivalent molecular volume with ratio of Mg:Fe" in talc is not known, and there are uncertainties about the crystal chemistry of talc. All the factors involved make it difficult to test the ideal of constant-volume control over composition.

The talc-carbonate rock--serpentinite boundary.-- The alteration of serpentinite to talc-carbonate rock took place at a moving boundary, grossly concentric with the borders of the ultramafic body, that marked the position at which the chemical potential of CO_2 attained a value such that serpentine was replaced by talc and magnesite; magnetite decreased in volume and probably altered in composition. As the reaction proceeded and the alteration was completed at successive positions, the boundary migrated inward toward the center of the ultramafic body. The principal features of the reaction were ingress of CO_2 and egress of H_2O. Other changes were limited essentially to adjustments of the various components among the minerals serpentine, talc, magnesite, and magnetite.

The serpentinite--dunite boundary.-- Theoretical considerations indicate that, concurrently with the talc-carbonate rock reaction, another moving boundary was defined by the conversion to serpentine (and magnetite) of any relict grains of olivine or masses of dunite in the ultramafic body, by reaction with water released from the talc-carbonate rock reaction. This boundary marked the position at which the chemical potential of H_2O attained a value such that olivine was unstable and was replaced by serpentine; it was concentric with the outlines of dunite masses and olivine grains. As serpentinization was completed at successive positions the boundary migrated inward toward the centers of the dunite masses. Ingress of H_2O and loss of Mg were the principal features of the reaction. The Mg may have migrated to the talc-carbonate rock zone to form magnesite. However, it has been shown above that the proportion of olivine and dunite in the ultramafic bodies here considered was probably very small at the beginning of steatitization; the amount of serpentine formed by such reaction was probably negligible.

The blackwall--steatite reaction

Within the limits of the system embraced by the blackwall and steatite, it is evident from table 1-D and figure 5 that steatitization involved chiefly the addition of Si and the loss of H_2O. Interchange between rocks within the system of a few other constituents was relatively great, but the gains and losses to the system of these and of the remaining constituents was small. By enlarging the system to include a few feet (possibly as much as 10 feet) of the schist bordering the blackwall, the gains and losses to the system could be reduced essentially to zero except for H_2O, which could then be considered the sole mobile constituent. (CO_2, though migrating freely through the blackwall--steatite zones, does not enter into the reactions in those zones in significant amounts, and plays a "transient" role.) In other words, the steatite--blackwall alteration can be regarded as a metamorphic-differentiation reaction (Phillips and Hess, 1936, p. 333-362) in which only H_2O is gained or lost to the system, if the added SiO_2 is regarded as being derived from the schist bordering the blackwall and if the other minor changes originate or terminate in the schist within a few inches or a few feet of the blackwall. This condition is obviously true for Na, which migrates outward only as far as the albite porphyroblast zone bordering the blackwall. The evidence suggests, but is not conclusive, that the same is true of K and biotite. The migration of Al is partly tied in with that of Na and the development of albite porphyroblasts, and is consistent with the conditions postulated. The condition for the other elements is less easily checked, but there appears to be no evidence against the model picture in any instance, and it may be reasonably assumed that they, too, behave accordingly. With

from schist rather than outside the system?

this model system as a basis for discussion, the blackwall--steatite reaction may be generalized about as follows:

Steatite and blackwall formed simultaneously by metamorphic differentiation, with the alteration of serpentinite to steatite supplying Mg and H_2O (the latter in excess) to the developing blackwall zone, and the alteration of schist to blackwall supplying Si to the developing steatite zone. Simultaneous alteration to steatite of the schist immediately next to the original contact of the serpentinite supplied Al to the developing blackwall zone. With continuation of the process the inner portion of the blackwall was altered to steatite, supplying Al to the continuously widening blackwall zone and deriving Si from the schists beyond; the steatite zone was also widening at the expense of serpentinite (at a much greater rate than at the expense of former schist), so that Mg was continuously supplied to the growing blackwall zone. Movement of other constituents was relatively unimportant, in most cases, and will be considered later under the section "Geochemistry of the rocks affected by steatitization."

Tremolite formed locally at the contact between blackwall and steatite where conditions, such as availability of Ca, were favorable. Physical conditions were very close to those of the lower limits at which tremolite is stable, so that the distribution of tremolite is spotty, the crystals comparatively small, and the amount distinctly limited. Progressive changes in bulk composition through addition of Mg from the serpentinite, loss of Si to the talc, and perhaps in loss of Ca to carbonate because of rising partial pressure of CO_2 with duration of metamorphism, resulted commonly in partial replacement of tremolite by talc.

The conspicuous albite porphyroblast zone at the outer border of the blackwall next to albitic schist formed concurrently with the blackwall as a result of the outward displacement of Na (and, probably, in some instances, of Al) by the advancing wave of chlorite of the blackwall, which replaced nearly all minerals in its path. The displaced Na (and Al) reacted almost immediately beyond the advancing front of blackwall with constituents of the schist, particularly quartz, to form heavy concentrations of albite porphyroblasts. Biotite and muscovite, and the associated K, probably behaved somewhat analogously to albite and the associated Na, but the data are not so conclusive nor the results so striking; they probably dispersed farther into the schist. Graphite appears to have been displaced by the chlorite of the blackwall as if "flushed out" so that it formed heavy concentrations at the contact between blackwall and schist.

Apatite, epidote, zircon, pyrite, ilmenite, sphene, and rutile were all relatively persistent during steatitization, but most were involved to some extent in the reactions. The general decrease, inward from the schist, of the content of apatite and epidote in the blackwall and outermost steatite, and the textural evidence of replacement shown by many of the grains, indicates that both were slowly but progressively replaced by chlorite and talc. Some of the P of apatite may have substituted for Si in chlorite and talc or all may have migrated outward. Ca from both apatite and epidote probably reacted with the titanium minerals in the blackwall zone in the manner discussed immediately below. The distribution of pyrite is so irregular that its behavior during steatitization cannot be accurately deduced; some grains appear to have been partially replaced by chlorite. Zircon appears to have persisted without being affected by any reactions. Ilmenite, rutile, and sphene show general relationships that indicate successive alteration of ilmenite ---> rutile ---> sphene. Both ilmenite and sphene are common in schist and greenstone, and laths of ilmenite are commonly bordered by rims of sphene, but the full sequence ilmenite ---> rutile ---> sphene was observed only in the blackwall. The alteration appears, therefore, to be associated in space and time with steatitization. There also appears to be a relative diminution of total titanium minerals at the inner border of the blackwall zone and in the steatite derived from schist.

The reactions among the titanium minerals could not have resulted primarily from changes in temperature, if the earlier conclusions on temperature relations during steatitization be correct (see p. 31-32); the reactions must, then, have occurred as a result of other factors, such as bulk chemical changes brought about by steatitization or the making available of elements released by other reactions. The alteration of ilmenite ($FeTiO_3$) to rutile (TiO_2) involves loss of FeO; the alteration of rutile to sphene ($CaTiSiO_5$) requires addition of CaO and SiO_2. These several restrictions suggest the following explanation for the reactions:

In the early stages of steatitization (or in the outer part of the blackwall zone) ilmenite altered to rutile because the blackwall, being composed entirely of chlorite, could accommodate much more iron than could the schist, and iron consequently diffused from the ilmenite to the chlorite. A similar reaction with biotite may have occurred where concentrations of biotite were formed just outside the blackwall. In the later stages of steatitization (or near the inner part of the blackwall zone), CaO released by the slow alteration of epidote and apatite -- and perhaps from calcite -- reacted with rutile and SiO_2 (migrating from the schist inward toward the steatite zone) to form sphene. Locally, agglomeration of finely dispersed grains about relatively few nuclei appears to have occurred. Throughout the steatitization process the total content of ilmenite, rutile, and sphene in the rocks being altered to blackwall and steatite was reduced proportionately to the extent that Ti could enter into the structure of chlorite and talc.

The diverse reactions and interchanges of material deduced for the blackwall-steatite alteration are best understood and most simply interrelated if they are considered in terms of reaction at moving boundaries, each marking the position at which the chemical potential of a mobile constituent attained a value critical to a significant mineral reaction. Four principal moving boundaries of this nature existed in the blackwall--steatite reaction.

<u>The steatite--serpentinite boundary</u>.-- The steatite--serpentinite boundary marked the position at which the chemical potential of Si attained a value such that serpentine (and magnetite) were unstable and were replaced by talc. Addition of Si, and diffusion of Mg outward toward the schist, and loss of H_2O were important features of the reaction. The boundary was essentially concentric with the outlines of the ultramafic body, and migrated slowly inward toward the center of the body as the alteration of serpentinite to talc was completed at successive positions.

<u>The steatite--talc-carbonate rock boundary</u>.-- At most places throughout the duration of the blackwall--steatite alteration, talc-carbonate rock bordered the steatite zone inwardly, and the reaction was between steatite and talc-carbonate rock. The boundary between the two marked the position at which the chemical potential of Si attained a value such that magnesite was no longer stable, and the talc-magnesite assemblage was replaced by a talc assemblage. Addition of Si and H_2O and loss of Mg and CO_2 were the main features of the reaction. The boundary was between the talc-carbonate rock--serpentinite boundary and the border of the ultramafic body. As the steatite alteration proceeded, the steatite--talc-carbonate rock boundary migrated inward toward the center of the body at a slower rate than the talc-carbonate rock--serpentinite boundary.

The blackwall--schist boundary.-- The blackwall--schist boundary was actually a narrow zone marking critical values in the chemical potentials of several components, principally Mg, Al, Si, Na, and K. In this zone, several reactions took place so close together in space as to be difficult to separate. Theoretically, successive reactions should have resulted in mineralogically distinct successive zones, but the smallness of the differences in chemical potential for the different components responsible for the successive reactions, and the slowness of the reactions, resulted in the telescoping of the different zones at the outer border of the blackwall zone. As critical values in the chemical potential of Mg and Al were reached, the minerals of the country rock -- principally quartz, sericite, chlorite, and albite -- were replaced by a monominerallic magnesian chlorite assemblage. The chief features of the reactions were the ingress of Mg and H_2O from the steatite--serpentinite (or steatite--talc-carbonate rock) boundary and of Al from the steatite--blackwall boundary; and displacement of Si, which migrated to the steatite--serpentinite (or steatite--talc-carbonate rock) boundary, and of K and Na, which moved outward toward the schist. The boundary marking the alteration of the minerals of the schist to chlorite migrated outward from the border of the ultramafic body into the schist as the reaction proceeded. Immediately in front of the advancing chlorite boundary, Na and K attained chemical potentials such that albite and biotite were formed.

Reactions involving apatite, epidote, zircon, pyrite, ilmenite, rutile, and sphene did not form well-defined boundaries, principally because the reactions were very slow. Some, such as the ilmenite --> rutile --> sphene reaction, appear to have resulted from local achievement of critical values in the chemical potentials of certain elements, such as Ca and Si, as a result of the alteration to chlorite of other minerals. All (except probably zircon and pyrite) appear to have altered slowly to chlorite under the mass effect of ingress of Mg, Al, and H_2O.

<u>The steatite--blackwall boundary</u>.-- The steatite--blackwall boundary marked the surface at which the chemical potential of Mg had a value such that chlorite was replaced by talc. The principal features of the reaction were the ingress of Mg from the steatite--serpentinite (or steatite--talc-carbonate rock) boundary, and the migration outward toward the schist of the displaced Al, and release of H_2O. The boundary moved progressively outward toward the schist at the expense of the blackwall, probably at a somewhat slower rate than the blackwall--schist boundary advanced. Most of the original minerals of the schist were entirely replaced by chlorite of the blackwall by the time that the steatite--blackwall boundary advanced to a given point, but locally, such persistent minerals as apatite, zircon, and sphene persisted for a short distance inside the steatite--blackwall boundary.

GEOCHEMISTRY OF THE ROCKS AFFECTED BY STEATITIZATION

The distribution of atoms, ions, and radicals among the rocks affected by steatitization is shown by, and their relative movement readily inferred from, the rock formulas in table 2-C. The inferred relative movement is, of course, based upon the premise that replacement was on a volume-for-volume basis. The relations between some of the mineral changes and the movements of some constituents are obvious upon comparison of the graphs showing the mineral composition of the rocks (fig. 4) and those showing chemical composition of the modified standard cells (fig. 5). For other constituents the relations are not so obvious. In the remainder of this section the geochemistry of each of the constituents on which information is available -- the mineral associations of elements in the original rocks, the course of movement of elements during steatitization, and the final disposition of the elements -- will be discussed briefly, and subject to the limitations of data that are in some instances rather spotty.

Potassium is confined, as an essential constituent, to the schist, where K_2O commonly forms as much as 3-4 percent by weight of the rock, and where it occurs in the minerals muscovite and biotite. All the other rocks contain generally less than 0.1 percent of K_2O; but all, even such rocks as serpentinite and steatite, in which no minerals ordinarily regarded as containing potassium are detectable microscopically, contain generally a few hundredths percent. The reported presence of K_2O in such small amounts may result from contamination, from analytical errors, or from the actual presence of minute amounts of potassium in substitution for major components. In the serpentinite, talc-carbonate rock, and steatite, analytical error or contamination would seem to be likely explanations. In the blackwall and greenstone, minute amounts of sericite or small amounts of amphibole may account for the K_2O, or small amounts of K may be accommodated within the chlorite structure, such as coupled with the substitution of Al for Si.

In the steatitization process K is removed almost entirely from the rock in the alteration of schist to blackwall and steatite. Local concentrations of biotite in the schist near the blackwall indicate that at least part of the K so displaced is stabilized almost immediately outside the blackwall zone. Probably all the K displaced comes to rest very near the blackwall zone, but the normal content of muscovite in the schist is so variable that such a conclusion must remain largely speculative.

Sodium plays a conspicuous role locally in the changes effected during steatitization. Sodium occurs in significant proportions only in albitic varieties of schist, in greenstone, and in the albite porphyroblast rock that borders the blackwall locally. Na_2O commonly forms as much as 1 to 3 percent by weight of albitic schist and greenstone, and 6 to 8 percent of albite porphyroblast rock. In other varieties of schist Na_2O commonly forms not more than 0.X to 0.0X percent (by weight) of the rock. The Na appears to be contained almost entirely in albite. Substitution of Na for K in muscovite appears to be slight at most, and paragonite is generally absent from the schist.

Sodium is removed from the rock in the alteration of schist to blackwall and steatite, and is concentrated at the outer margin of the blackwall zone. The process involves the disintegration of albite within the blackwall zone and the deposition of albite in the albite porphyroblast zone, but the outward migration probably involves only Na, which reacts immediately beyond the influence of the advancing wave of chlorite with the requisite constituents to form albite, whereas the Si released from the albite diffuses toward the steatite, and the Al toward the blackwall or toward the albite porphyroblast zone, depending upon chemical requirements.

Calcium is an essential and generally major constituent of thin beds of sedimentary carbonate rock in the schist, of actinolitic greenstone, and of actinolite rock (between the blackwall and steatite); it is variable in proportion but generally rather abundant in chloritic greenstone; and it generally forms less than 1 percent (weight percent CaO) of the schist and 0.0X percent of blackwall, steatite, talc-carbonate rock, and serpentinite. In the sedimentary carbonate beds the Ca occurs in the mineral dolomite. In schist, greenstone, and actinolite rocks, the Ca is present in such calcian minerals as epidote, sphene, apatite, amphibole, and carbonate. In blackwall the generally very low content of Ca may possibly be accommodated partly within the chlorite structure, but most probably occurs in relict minerals such as epidote and amphibole, and in sphene; where the CaO content of the blackwall is much greater than 0.X percent, the presence of local segregations of epidote or sphene is generally indicated; in a few instances calcite rhombs that have formed by replacement after steatitization are responsible for a relatively high content of calcium. In steatite, talc-carbonate rock, and serpentinite, the calcium appears to be contained almost entirely within carbonate, chiefly as a minor constituent of magnesite but in a few places in small amounts of dolomite.

During the steatitization process, Ca is released during the alteration of schist or greenstone to blackwall. A small part of the Ca released remains in the blackwall zone, entering into the chlorite structure or taking part in the reactions of the titanium minerals discussed above. The rest probably migrates to the talc-carbonate rock zone and enters into the composition of the carbonate being formed there.

Magnesium is a persistent constituent of all the rocks and is a major component of the ultramafic rocks and those related to steatitization. The schist commonly contains less than 5 percent (by weight) of MgO, except where chlorite is very abundant. Beds of sedimentary carbonate rock (dolomite) are highly variable and contain 20-40 percent MgO. Serpentinite contains about 37-40 percent MgO, talc-carbonate rock about 30 percent, steatite about 29 percent, and blackwall 15-25 percent. Greenstone ranges in content of MgO from about 10 to 20 percent. Actinolite rock contains about 20 percent MgO. The magnesium is contained almost entirely in serpentine, talc, and carbonate in the ultramafic rocks, and in chlorite, amphibole, and biotite in the other rocks. Some Mg is probably contained in magnetite, particularly within the serpentinite and talc-carbonate rock, but the proportion of Mg to Fe is probably small.

Mg was immobile in the alteration of serpentinite to talc-carbonate rock, but was displaced during the alteration of serpentinite to steatite and migrated outward into the immediately adjacent schist. The chlorite of the blackwall, which formed in the adjacent schist concurrently with the steatite, took up the Mg displaced by the alteration of serpentinite to steatite.

all?

Both ferrous and ferric iron are persistent constituents of all the rocks. Ferrous iron is a major constituent of the blackwall and greenstone, of which it constitutes 10-20 percent by weight of FeO; it commonly forms about 5 percent of actinolitic greenstone, talc-carbonate rock, steatite, and serpentinite, and 1-2 percent or less of the schist. Ferric iron seldom forms more than 0.X percent of any of the rocks except for serpentinite, and locally talc-carbonate rock, which commonly contain 3 percent or more Fe_2O_3.

The Fe'' in all the rocks occurs principally in silicate minerals and in carbonate, in limited isomorphous substitution for Mg. The chief silicates that contain appreciable Fe'' are serpentine (antigorite), chlorite, actinolitic hornblende and actinolite, and talc. Among the carbonates, most of the Fe'' occurs in magnesite, but some also occurs in dolomite. Magnetite contributes appreciable amounts of Fe'' in most serpentinite and talc-carbonate rock, and locally in sedimentary carbonate beds and in greenstone; some steatite contains small amounts of Fe'' in dustlike particles of magnetite. Ilmenite contributes a small percentage of Fe'' to much of the schist.

Fe''' contained in magnetite is distributed among the different rocks as described above for Fe'', and accounts for most of the Fe''' in serpentinite, talc-carbonate rock, steatite, and carbonate rock. Greenstone, and some schist, contain a relatively large proportion of total Fe''' in epidote, in which the Fe''' substitutes for Al. Other rocks contain Fe''' chiefly in chlorite and biotite, in substitution for Mg, coupled with the substitution of Al for Si.

In the steatitization process it seems clear that Fe" is lost in the alteration of serpentinite to steatite, and that part of the Fe" thus displaced is concentrated in the chlorite in the blackwall zone. The evidence for gain or loss of Fe" and Fe'" in the other zones is uncertain, chiefly because of the variable distribution of magnetite within serpentinite and talc-carbonate rock. The evidence suggests that total Fe remained about constant in the talc-carbonate rock reaction, and that Fe" increased slightly at the expense of Fe'" (see table 2-C and fig. 5) by reduction of Fe'" in magnetite. Ingress of very small amounts of Fe" from the steatite zone into the talc-carbonate rock may have occurred in places, and Fe" may have been introduced rarely from outside the ultramafic body.

Manganese is a minor but persistent constituent of all the rocks. MnO commonly constitutes 0.X percent by weight of all the rocks except serpentinite and steatite, of which it generally makes up only 0.0X percent. The Mn is similar in chemical properties and in atomic radius to Fe", and the content of Mn shows a close relation, in most instances, to the proportion of Fe". Consequently, Mn is distributed in a manner closely analogous to that of Fe", but the carbonate minerals consistently contain a higher ratio of Mn/Fe" than do the silicates. The movement of Mn during steatitization was probably very similar to that of Fe".

Nickel, though ubiquitous, is always a very minor constituent, but is clearly more abundant in rocks derived from ultramafic igneous rocks than in those derived from sedimentary rocks. Serpentinite, talc-carbonate rock, and steatite commonly contain 0.X percent of NiO by weight, whereas schist and blackwall contain 0.0X percent. Carbonate formed during serpentinization and steatitization contains about ten times as much Ni as that in the late talc-carbonate veins: carbonate in talc-carbonate rock and serpentinite contains 0.0X percent Ni, whereas that in the younger late veins contains 0.00X percent. Some Ni in the ultramafic rocks is contained in sulfarsenide minerals, but most occurs in silicates and in carbonates in substitution for Mg and Fe". The Ni in the schist and blackwall presumably occurs in chlorite in similar substitution. No changes in content of Ni as the result of steatitization is evident in any of the rocks.

Cobalt occurs uniformly in small amounts in all the rocks. No consistent differen~~t~~ce in Co content is apparent between the ultramafic igneous rocks and the rocks derived from sedimentary rocks; in all, CoO constitutes about 0.0X weight percent of each rock. Mineral analyses of talc and carbonate show that the Co is contained partly in those minerals, but that the proportion is lower than in the rock as a whole, so that other minerals must also contain appreciable Co. Inasmuch as cobalt is a member of the iron family, the balance of the Co in the rocks probably occurs in silicate minerals in substitution for Mg in the same manner as Fe" and Mn. The Co content of the rocks shows no evidence of change during the steatitization process.

Aluminum is a major constituent of the rocks of sedimentary origin, of which Al_2O_3 commonly forms 15-25 percent by weight; but it is a minor constituent of the ultramafic rocks, of which Al_2O_3 forms only about 1 percent or less. In the rocks of sedimentary derivation, aluminum is associated principally with the silicates muscovite, biotite, chlorite, epidote, albite, and almandite. In the ultramafic rocks the small amounts of Al occur both in the silicates (talc and antigorite)~~,~~ and in magnetite. In talc and antigorite the Al is generally considered to substitute entirely or almost entirely for Mg. In epidote and garnet, Al is a stoichiometric constituent. In the other silicates the Al is in coupled substitution, in fourfold and sixfold coordination positions, for silica and a divalent metal ion.

As for Fe" and Fe'", there is some uncertainty about the relative movement of Al during steatitization, because of the irregular distribution of magnetite in serpentinite and talc-carbonate rock. The alteration of serpentinite to talc-carbonate rock and steatite seems generally to have resulted in a diminution of Al. The Al so displaced migrated outward to enter into the chlorite in the blackwall reaction, or into the albite of the albite porphyroblast zone. The Al content of the blackwall zone increased or diminished with respect to that of the original schist depending upon whether the original schist was nonalbitic or albitic.

Chromium occurs in small amounts in all the rocks but is appreciably more abundant in the ultramafic rocks, where Cr_2O_3 forms 0.X percent by weight, than in those of sedimentary derivation, where Cr_2O_3 forms 0.0X percent. Chromium occurs in the serpentinite and talc-carbonate rock chiefly in chromian magnetite, but in a few places occurs in translucent grains of chromite; both talc and serpentine also contain small amounts of Cr. In the metamorphosed sedimentary rocks (including the blackwall), the Cr presumably is contained entirely in the silicates such as chlorite and mica. No appreciable movement of Cr appears to have occurred during steatitization.

Titanium occurs in both the metamorphosed sedimentary and volcanic rocks and in the ultramafic rocks, but is generally ten- to several hundred-fold more abundant in the former. In most schist and greenstone TiO_2 forms 1 percent by weight or less of the rock, but in a few places it runs as high as 3-5 percent. The ultramafic rocks invariably contain no more than 0.0X percent TiO_2.

In the schist, greenstone, and blackwall, the Ti is contained principally in the minerals ilmenite, sphene, and rutile; small amounts probably occur also in chlorite and biotite. The Ti in ultramafic rocks presumably occurs in the talc and antigorite.

Movement of Ti during steatitization appears to have been slight. The reactions involving ilmenite, rutile, and sphene in the blackwall have been discussed. The end stages of chloritization in the blackwall and the succeeding alteration of the innermost shell of blackwall to steatite appears to have displaced Ti outward.

Silicon is a major component of all the rocks except the purer beds of sedimentary carbonate. The content of SiO_2 ranges from 0.X to X. percent by weight in the carbonate beds, through 30-40 percent in the serpentinite, talc-carbonate rock, greenstone, and blackwall, to about 60 percent in the schist and steatite; many relatively thin beds of quartzite are nearly 100 percent SiO_2. The principal silicates with which the Si is associated are quartz, muscovite, chlorite, albite, amphibole, talc, serpentine, epidote, and sphene.

The alteration of serpentinite to talc-carbonate rock requires little or no movement of Si, but the blackwall--steatite reaction resulted in addition of Si to the steatite zone and loss of Si from the blackwall zone. The blackwall zone could supply only about one-third, generally, of the Si necessary for the steatite zone; the balance was contributed principally in small increments from throughout the schist adjacent to the ultramafic body.

any real evidence or just an available source? [illegible]

Phosphorus occurs in very small amounts in all the rocks. The content of P_2O_5 in the metamorphosed sedimentary rocks is about 0.1-0.2 percent, and in the ultramafic rocks is 0.01-0.02 percent. The P in the rocks of sedimentary derivation is associated in all cases with apatite. No apatite was observed in steatite, talc-carbonate rock, or serpentinite. Presumably, therefore, the P in the ultramafic rocks occurs in silicates in the Si position. Movement of P in steatitization was very slight. Apatite was very slowly replaced in the alteration of schist to blackwall and steatite; the very small amounts of P thus displaced may have remained essentially immobile, substituting for Si in either chlorite or talc.

Carbon as CO_3 anions in carbonate is a major component of talc-carbonate rock and of sedimentary carbonate beds, and is a common minor constituent of the other rocks. Free carbon, probably both amorphous and as crypto-crystalline graphite, occurs only in varieties of schist, in quartzite, and in albite porphyroblast rock. In the alteration of schist to blackwall and steatite, C (free) was displaced from the schist in the alteration to chlorite and was flushed ahead of the advancing wave so that heavy concentrations of graphite were formed at the outer margin of the blackwall zone. Locally, residual enrichment may have been a factor in the concentration, but was not generally important. Carbon as CO_2 was highly mobile during steatitization; its introduction into the serpentinite was the most important factor in the alteration of serpentinite to talc-carbonate rock.

Hydrogen, in water or OH, is an essential constituent of all but the carbonate rocks and quartzite. The content of H_2O ranges from less than 2 percent by weight for some varieties of schist to nearly 12 percent in blackwall and serpentinite. The anion OH occurs in the silicates chlorite, serpentine, muscovite, biotite, amphibole, and epidote; and in apatite. Water was strikingly mobile in the steatitization process. The alteration of serpentinite to talc-carbonate rock and steatite resulted in the loss of much water, part of which was taken up in the blackwall by the alteration to chlorite, and part of which was expelled entirely from the system.

Oxygen is, of course, a principal constituent of all the rocks and all the minerals except the sulfides and sulfarsenides, in terms both of molecular percent, weight percent, and volume percent, but particularly in terms of volume percent. On a volume basis, oxygen makes up about 94 percent (of the ion volume) of most rocks (Barth, 1952, p. 82). The movement of oxygen during steatitization was tied in primarily with movement of H_2O and CO_2, and the gains and losses of total oxygen in most rocks was relatively small. Small movement of oxygen appears to have been tied in with changes in valence state of metallic ions, particularly Fe" and Fe'". In blackwall and steatite derived from schist, the relative change in content of oxygen with respect to the content of the parent rock varies considerably from place to place because of differences in the parent rock. On the other hand, talc-carbonate rock and steatite derived from serpentinite show consistent changes in total content with respect to the parent rock because the serpentinite is generally uniform. In both, and particularly in the talc-carbonate rock, total oxygen per modified standard cell increases appreciably.

Sulfur is a common minor constituent of all the rocks but is erratically distributed. The S content of schist varies from about 0.0 to 0.X percent by weight; the ultramafic rocks generally contain not more than 0.1-0.2 percent, but in a few instances contain 0.5 percent or more. The S in the schist is contained entirely in pyrite. In the ultramafic rocks it is contained in pyrite, pyrrhotite, and sulfarsenides such as gersdorffite.

The distribution of arsenic is partially known only for the ultramafic rocks and blackwall. The content of As_2O_3 was 0.0003 weight percent in a single specimen of blackwall, and 0.0009-0.065 percent in four samples of ultramafic rock. The As occurs in sulfarsenides, such as gersdorffite.

much water must be lost - and must get out by least means. Why not these contacts? At green schist temperature, this water could easily have contained 500± ppm SiO_2, accounting for steatite. Such water is "metamorphic" and not magmatic. DEW

NATURE OF THE STEATITIZING "SOLUTIONS"

The discussion in the foregoing section clearly leads to the conclusion that in steatitization the formation of talc-carbonate rock from serpentinite requires only the introduction of CO_2, with the resulting expulsion of considerable H_2O. The formation of steatite requires the addition of Si, but that has been shown to be readily accounted for as being derived principally from the blackwall zone and the adjacent schist, through diffusion during metamorphic differentiation. It remains, therefore, only to account for the source of CO_2. Obviously, then, the "solutions" can be of extremely simple nature; they may have consisted almost entirely of CO_2. Locally, they appear to have transported small amounts of silica.

Possible but not "shown"

Most investigators ascribe steatitization to dilute hot aqueous solutions (presumably rich in CO_2) derived from nearby or underlying acid igneous rocks. The close connection shown to exist between regional metamorphism and steatitization suggests, however, that the CO_2 may simply have been derived from the rocks of sedimentary parentage during progressive metamorphism. A common reaction during progressive metamorphism that would lead to release of CO_2 is the reaction between carbonate and silica or silicates to form lime and magnesian silicates and CO_2. Another possible source is suggested by the apparent antipathy between magnetite and graphite in regionally metamorphosed rocks, which suggests that hematite and free carbon react to form magnetite and CO_2. Dependence upon such reactions for a source of CO_2 is not required exclusively, of course, if regional metamorphism is attributed to magmatic effects, nor is such dependence ruled out; indeed, under such conditions, a mixed source for the CO_2 (that is, a source both in the sediments and in the magma) seems likely. But the evidence of magmatic control of metamorphism in the area is less than compelling, and it seems likely that the "solutions" that effected steatitization were independent of a magmatic source.

ORIGIN OF THE TALC-CARBONATE VEINS

The talc-carbonate veins are clearly later than and unrelated to steatitization, as shown by the facts that they crosscut the talc-carbonate rock, steatite, and serpentinite, and that the coarse carbonate and talc are essentially undeformed. Their exact age cannot be determined, but the veins are obviously related to late joints, which may have formed as late as the joints along which were admitted the diabase dikes of probable Permian or Triassic age.

No talc-carbonate veins were observed to extend beyond the ultramafic bodies, but none was exposed sufficiently so that it could be proved that a given vein did not extend into the country rock. The fact that no talc-carbonate veins were observed within the blackwall in thousands of feet of exposure is strong evidence that the veins are confined to the ultramafic rocks. Only in the case of Ca does it appear necessary to call upon introduction from an outside source, and that may well have been the immediately adjacent schist. The veins are clearly joint controlled, but were formed in considerable part by replacement, as is attested by the irregular boundaries. In contrast to structural conditions during steatitization (see p. 27), the joints probably formed relatively open channelways, and water probably pervaded all joints, fractures, and grain-boundary spaces. Mg, Si, CO_2 (or CO_3), and Fe" could readily have diffused into the veins from the bordering ultramafic rocks. Ca was introduced along the joints either by mechanical movement of material in solution or by diffusion through the solution. The veins grew probably both by deposition in open fissures (probably essentially simultaneously with opening of the fissures), and by replacement of adjoining steatite, talc-carbonate rock, or serpentinite.

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