

THE WATCHUNG BASALT AND THE PARAGENESIS OF ITS ZEOLITES AND OTHER SECONDARY MINERALS

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INTRODUCTION

Certain localities within the area covered by the basaltic ridge which forms the First Watchung Mountain in Northern New Jersey have for many years been famous among mineral collectors in the East for the beautiful specimens of zeolitic minerals which they have yielded. Although from a purely utilitarian point of view nothing of economic value has been discovered among these minerals or their associates, yet to the mineralogist, the size and perfection of crystal groups and the delicacy of form and coloring which are exhibited render them most attractive and desirable acquisitions. In these respects, the finest specimens found in the New Jersey localities are but little inferior to the similar zeolitic groups brought from the world-famous collecting-grounds at Berufjord in Iceland and the Poonah District in India.

Beyond the attraction which these minerals offer as objects of beauty or from a mineralogic standpoint, they present to the geologist many problems which add greatly to their interest. From this side, the relations which they bear to the enclosing rock and the cause and manner of their deposition are matters of greater importance. It is easy to recognize from their field associations that the veins and pockets in which they are found have many points of similarity with metalliferous veins, and that a full understanding of the processes involved in the formation of the zeolites would be of the greatest value in interpreting certain of the features of the economically important metalliferous deposits. From another point of view, one can recognize in the genesis of the zeolites a phase of manifestation of the processes of hydrothermal metamorphism, which operate in various forms to alter the crystalline character and the mineralogic composition of igneous and sedimentary rocks.

Study of the zeolites by various workers has resulted in considerable literature on the subject. The greater part of what has been written, however, deals purely with the crystallographic or optic properties of the

minerals. It has been found that the optic characteristics present many anomalies, not all of which have found explanation, and that the laws governing the crystallographic forms of the various species are very complex, especially as regards twinning. These are matters which are largely of a physical nature and are apart from the scope of the present paper. They will not be considered, except that the results which have been attained will be utilized as offering valuable criteria for determination and discrimination. Efforts have also been made to determine the order of succession of the various associated species from different localities, but, so far as the writer is aware, the resources of microscopic petrography have been little utilized in this direction.

The present paper embodies the results of a study of various questions of geology and manner of formation of the zeolites and associated secondary minerals which are found within the area of Watchung basalt mentioned. The work has been carried out under the direction of the Geological Department of Columbia University, and the microscopic studies of rock sections have been made in the petrographic laboratory of that institution. The writer takes the greatest pleasure in acknowledging his sense of deep obligation to the members of the Geological Department, Professor Kemp, Professor Berkey and Professor Grabau, for their kindness and assistance throughout in guidance and suggestions.

The chief features which will be considered in the paper are the geologic conditions which gave rise to the formation of the secondary minerals and the order of succession of the various species as determined by petrographic methods, and in connection with this the results attained will be interpreted, so far as has been found possible, in the light of the laws of physical chemistry. The application of this branch of science in its modern aspects to the problems of the mineralogist and petrographer is at the present time at a most interesting stage, where the applicability and value of the principles are perceived, but application to specific cases has been slight.

I GEOLOGY OF THE REGION

CONDITIONS OF TRIASSIC DEPOSITION

An understanding of the major features of the geologic history of the region under discussion for a short period prior to the extrusion of the First Watchung basalt is essential for a comprehension of the processes which resulted in the formation of the secondary minerals. There has been some lack of agreement among different observers as to the interpretation of certain features of Triassic history. Earlier views were influ-

enced by the supposed necessity of attributing the accumulation of all sedimentary terranes to deposition in large bodies of water. The possibilities of fluvial accumulation on piedmont slopes or in depressed basins were not recognized. Later studies of the eastern Trias, carried out with this conception in mind, have shown that many strong arguments may be advanced favoring this interpretation, and there is now a strong tendency toward a general adoption of this point of view.¹ To the writer, the evidences in support of this view appear of great weight, and throughout the present paper it is accepted without reservation.

With this explanation, the conditions of Triassic deposition may be summarized as follows: The processes of mountain-folding which brought the Permian period to a close in eastern North America found expression along lines approximately parallel to those which had characterized previous periods of orogenic movement along the Atlantic border and resulted in the uplift of the Appalachians along axes having in general a northeasterly-southwesterly trend, but in places (as through Pennsylvania) departing quite widely from the general course. As a phenomenon which was probably closely associated with the structural instability attendant upon this period of orogenic development, secondary adjustments continued throughout a prolonged period.² The most important later movements of this character, so far as their history may be read, resulted in the formation of depressed areas closely parallel to the axes of Appalachian folding but in general somewhat to the southeastward of the main ridges. The downthrow or downwarping of these areas does not seem to have been sufficient to bring the depressions beneath the level of the sea, but, owing to their low-lying position, they formed, during the Triassic, areas of rapid accumulation of sediments of continental type, brought in by torrential streams as a result of the waste of the bordering uplands under climatic conditions of semi-aridity.

There is some question as to the former extent of the areas on which

¹ For evidences of sub-aërial accumulation see

W. M. DAVIS: 18th Ann. Rept. U. S. Geol. Surv., pt. 2, p. 32, 1896-1897.

J. V. LEWIS: "The Origin and Relation of the Newark Rocks," Ann. Rept. State Geol. N. J. for 1906, pp. 99-129.

C. N. FENNER: "Features Indicative of Physiographic Conditions Prevailing at the Time of the Trap Extrusions in New Jersey," Jour. Geol., vol. 16, pp. 229-327, 1908.

² I. C. RUSSELL: "The Newark System," Bull. 85, U. S. Geol. Surv., pp. 77 and 83, 1892.

E. J. SCHMITZ: "The Structure of the Richmond Coal-Basin," Trans. Am. Inst. Min. Eng., vol. 24, pp. 397-408, 1894.

J. V. LEWIS: *Op. cit.*, p. 107.

O. J. HEINRICH: "The Mesozoic Formation in Virginia," Trans. Am. Inst. Min. Eng. vol. 6, p. 266, 1878.

W. M. DAVIS: "The Triassic Formation of Connecticut," 18th Ann. Rept. U. S. Geol. Surv., pt. 2, p. 35, 1896-1897.

the sediments accumulated. On the one hand, it is contended that they were of wide-spread distribution and that the present more or less detached areas are merely remnants of a broad terrane. Others have argued in favor of local basins or troughs of depression, which may have presented analogies to the "graben" of the Rhine or to the Rift Valley of East Africa. To the writer, the intermediate view presented by the New Jersey Survey appears to offer many grounds for favorable consideration. It is as follows:³

These conditions are believed to be fully met by the hypothesis of river deposition across a relatively smooth Piedmont plain, fronting the newly uplifted crystalline foreland, or protaxis, of the Appalachian Mountains, with concurrent synclinal wrinkling and down-faulting of the long basin-like areas in which the present remnants of these rocks have been preserved. . . . Numerous short but vigorous streams brought down the débris of the disintegrating and decomposing granites, gneisses and metamorphic sediments of earlier Paleozoic age and deposited them in coalescing alluvial fans across the smoother plain of the crystalline Piedmont. Occasional downward movements, of warping or faulting, gave opportunity for local thickening of the deposits along the belts affected.

According to this conception, the Piedmont plain was of much greater width than the present areas of the Newark strata, and it probably merged into coastal marine and estuarine deposits along the eastern border. As a rule, however, the deposits were probably not very thick except in the elongated areas of progressive or intermittent deformation. Hence, when the whole Piedmont was eventually uplifted, the relatively thin mantle of débris was removed by erosion from the greater part of the region, and only those narrow belts that were protected by downwarping and faulting between adjacent areas of the harder crystalline rocks have been preserved to the present time.

The writer is inclined to lay especial stress upon the movements of deformation, which appear to have been renewed at intervals through a prolonged period at the same time that deposition was proceeding. They may have been a consequence, in part, of accumulating load, but they were also probably due to deep-seated causes connected with the primary movements. After each movement, the streams were rejuvenated, and the topographic depressions which otherwise would have been eliminated by the constant degradation of the bordering uplands and the accumulation of sediments were accented afresh.

Within these areas of fluvial aggradation, certain depressed portions of the surface (in New Jersey at least) were occupied by a series of shallow lakes when the First Watchung lava-flow occurred.* The existence of these lakes and their arrangement along lines parallel to the bordering

* J. V. LEWIS: *Op. cit.*, p. 107

* C. N. FENNER: *Op. cit.*, p. 309 a

highlands support the view that the movements of deformation caused the areas of accumulation to assume at times troughlike characteristics.

The lakes are believed to have been an all-important factor in giving rise to the conditions which resulted in the subsequent alteration of the basalt and the formation of the zeolitic minerals. Indications of their existence beneath the First Watchung flow can be found at Paterson and for a number of miles southward, and again near Plainfield.

IGNEOUS ACTIVITY

During some portion of Triassic time, probably the middle or later portion, the general uneasiness which had manifested itself in repeated crustal deformation assumed a somewhat different phase of expression. Numerous outbreaks of igneous activity occurred throughout the entire region from Nova Scotia to North Carolina. There is little indication that these outbursts anywhere assumed the form of centralized volcanic activity, but the fused material appears to have been ejected quietly through fissures whose direction coincides closely with the dominant lines of weakness. "It seems safe to conclude that both the dikes and faults are closely related and were probably, in part at least, contemporaneous."⁵

The dikes and extruded sheets are found not only in the areas now covered by Triassic sediments, but petrographically similar dikes have been traced through the intervening areas, and the region traversed by them is prolonged southward through South Carolina and Georgia and into Alabama, where they disappear beneath younger strata. The known length is about 1,000 miles. On the borders of the Triassic also, they occur in approximately parallel lines. The various intrusions are regarded by Russell⁶ as probably referable to the same general period, and the evidence appears to indicate that an area parallel with Appalachian folding, 1,000 miles or more in length and attaining a maximum width of 200 miles, was involved in a history of deformation and igneous injection which presents somewhat similar features throughout.

The chemical and mineralogical composition of the erupted magmas shows a remarkable uniformity.⁷ The general coincidence of the areas of eruption with the areas of deformation, and the extrusion of magmas which may be referred to closely related petrographic types, have undoubtedly a profound significance, but only the more superficial aspects can be interpreted.

⁵ I. C. RUSSELL, *Op. cit.*, p. 11.

⁶ *Op. cit.*, pp. 71-72.

⁷ E. S. DANA, *Am. Jour. Sci.*, 3d ser., 10, 1, 1892, 1894.

In Northern New Jersey, the igneous rocks associated with the Triassic (Newark) sediments include the intruded sheet of the Palisade diabase and the three extruded flows of Watchung basalt. The Palisade diabase appears to form a continuous sill, approximately parallel to the stratification, stretching diagonally across the State from New York into Pennsylvania but concealed in places by late sediments or glacial drift. The Palisade ridge in the North and Rocky Hill and Sourland Mountain in the South are considered portions of the same sheet. Its thickness is everywhere several hundred feet and in places is believed to reach nearly 1,000 feet.⁸ At several places, dikes and apophyses thrown off from the main mass reach the surface in small detached areas.

The three surface flows form the Watchung ridges and several isolated knobs. A maximum development of 1,200 feet is believed to be attained in the thickest parts of Second Mountain. In both First and Second Mountains, certain slight differences in composition and appearance vertically indicate a composite character of flow. An interbedded shale in the southern portion of Second Mountain is held by Lewis⁹ to be evidence of differential downward movements within the area between successive flows. Such movements would not be unexpected in a region experiencing the history described.¹⁰

LOCAL PECULIARITIES OF THE FIRST WATCHUNG SHEET

The basal layer of the First Watchung sheet apparently marks the initiation of igneous activity. The flow spread over the surface of the accumulating sediments and filled the depressions occupied by the playa lakes. Over the dry areas, the lava came to rest quietly, and the normal process of cooling and crystallization followed without interruption. The rock formed under these conditions presents a dense, homogeneous mass, which, under the microscope, is found to have the holocrystalline texture normal to basalts. Over the lake beds, however, the lava, which appears to have approached in composition the eutectic ratio of its component minerals and to have been almost at the temperature of crystallization thus determined,¹¹ was quickly chilled from the effects of water and vaporized steam. An exposure of the bottom layer in a quarry in the southwestern part of Paterson shows a mixture of lava and very fine mud

⁸ J. V. LEWIS: *Ann. Rept. State Geol. N. J.* for 1907, p. 129.

⁹ *Ann. Rept. State Geol. N. J.* for 1906, pp. 110-115.

¹⁰ Compare the geological history of the Rift Valley of East Africa as described by J. W. GREGORY: "The Great Rift Valley," London, 1896, especially chapter 12.

¹¹ C. N. FENNER: "The Crystallization of a Basaltic Magma from the Standpoint of Physical Chemistry," *Am. Jour. Sci.*, 4th ser., vol. 20, pp. 217-234, 1910.

several feet in thickness, and in many other places the lava was rendered extremely vesicular. The upper portions were not so thoroughly impregnated with steam but were, nevertheless, quickly cooled and became viscous. The jets and tongues of fused material seem to have assumed the consistency of a thick syrup and instead of spreading laterally they solidified in smoothly rounded boulder-like masses, having considerable similarity to the "pahoehoe" of Hawaiian flows which were first reported by J. D. Dana¹² and more fully described and illustrated by C. E. Dutton.¹³ The pasty character of the fluid and its sluggish movements are well attested in the billowy forms presented when quarrying operations have attacked bodies of trap of this character (Plate X). The rounded forms are sometimes built up to a thickness of 60-70 feet. The interior



FIG. 1. Microlitic growth of phenocrysts in vitrophyric crusts. $\times 35$. Slide 132.

of the boulders cooled with sufficient slowness to permit the basalt to crystallize with normal texture, but each is sheathed with a crust of glass (tachylite) varying from an inch to several inches in thickness, having often a laminated structure.¹⁴ Where unaltered, the color is usually dark olive-green or brown, and the appearance is decidedly vitreous. Under the microscope the rock is found to consist of a paste of glass, in which are set a few phenocrysts of labradorite and diopside, commonly surrounded by aggregates of hairlike microlites, which mark the continuation of growth of the phenocrysts during the initial stages of chilling before crystal-growth was wholly blocked by increasing viscosity (fig. 1).

¹² U. S. Exploring Expedition. Geology, p. 162.

¹³ 4th Ann. Rept. U. S. Geol. Surv., pp. 96 and 98, 1882-1883.

¹⁴ B. K. EMERSON: "Labase Pitchstone and Mud Enclosures of the Labase Trap of New England," Bull. Geol. Soc. Am., vol. 8, pp. 59-86, 1897.

Between the glassy crusts and the holocrystalline interior, transitions of crystal-development produce aphanitic phases.

The crusts frequently present a shattered appearance due to the sudden chill which they experienced, and at times pockets among the boulders are filled with considerable masses of breccia of this nature. This was probably originally a usual feature, but, as will appear presently, such pockets of glass were, for several reasons, peculiarly subject to the action of the processes which formed the secondary minerals.

The chemical composition of the basalt is shown in the following analyses of typical examples:¹⁶

	I	II	III	IV
SiO ₂	51.78	51.36	51.82	50.19
Al ₂ O ₃	12.79	16.25	14.18	14.65
Fe ₂ O ₃	3.59	2.14	0.57	3.41
FeO	8.25	8.24	9.07	6.96
MnO	0.44	0.09	0.13	0.07
MgO	7.63	7.97	8.39	7.95
CaO	10.70	10.27	8.60	9.33
Na ₂ O	2.14	1.54	2.79	2.64
K ₂ O	0.39	1.06	1.26	0.75
TiO ₂	1.41	1.17	1.13
P ₂ O ₅	0.14	0.17	0.18
H ₂ O	{ 0.63 }	{ 1.33 }	1.70	3.04
CO ₂				

Analysis I was recast by Hawes to show percentage of mineral constituents as follows:

Anorthite, 15.52; albite, 22.16; potash feldspar, 2.32; augite, 54.47; titanite iron, 2.68; magnetite, 1.76; apatite, 0.32; total, 99.23.

The augite was isolated by specific gravity solutions and found to have the following composition:

SiO ₂	50.71
Al ₂ O ₃	3.55
FeO	15.30
MnO	0.81
CaO	13.35
MgO	13.63
Na ₂ O {	
K ₂ O {	1.48
Loss on ignition	1.17

¹⁶ I. West Rock, New Haven, Conn. G. S. Analyst. Analyst. Rep. U. S. Geol. Surv. 4, pp. 129-134, 1881.

II. Watchung Mt., N. J. I. G. Eakins. Analyst. Bull. 150, U. S. Geol. Surv. p. 255, 1898.

III. Scotch Plains, N. J. R. B. Goss. Analyst. Ann. Rept. State Geol. of N. J. for 1907.

IV. Nr. Springfield, N. J. R. B. Goss. Analyst. Ann. Rept. State Geol. of N. J. for 1907.

Petrographic examination of the unaltered basalt in the area with which we are more especially concerned accords with the above analyses and shows a rock of simple mineralogic composition. The components are a pyroxene approaching diopside in optic properties, but shown by a large extinction-angle to deviate somewhat from the typical mineral; a plagioclase feldspar of the composition of labradorite; and magnetite. The texture is normal ophitic. The appearance is illustrated in Plate XI, fig. 1.

In both the crystalline and glassy facies, remnants of phenocrysts of resorbed olivine are not uncommon. These phenocrysts appear to have been developed during a period of intra-telluric crystallization and to have become unstable as a consequence of changes in temperature and

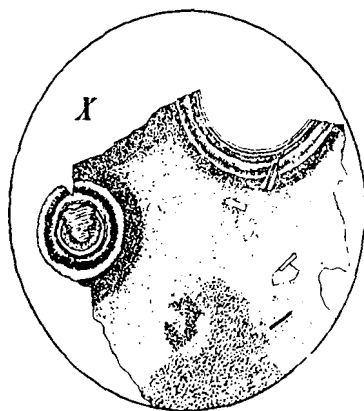


FIG. 2. Chloritic nodules from resorption of olivine (X = open field). $\times 35$
Slide 48.

pressure attendant upon extrusion, with simultaneous variation in the composition of the magma consequent upon the evolution of contained vapors. Resorption of olivine by the magma often proceeded so far that the only traces of the mineral that survive are small, spheroidal nodules, which, by subsequent alteration, have been converted almost wholly into chlorite or serpentine (fig. 2). The general character of the crystallized rock and such special features as the resorption of olivine are of importance in following out the process of formation of the secondary minerals; for vestiges of the primary character often remain even in advanced stages of alteration and show clearly that much of the action of recrystallization proceeded from stage to stage in a close-textured rock, without the presence of any important cavities.

A study of the field relations has shown that the spheroidal masses of

basalt, sheathed with glassy crusts, appear in those areas which, at the time of the lava flow, were covered by shallow lakes, and that these areas are coincident with those in which the formation of zeolitic minerals has occurred.

Evidence along various lines leads to the belief that the processes of alteration were directed by the features impressed upon the solidified basalt by the presence of the bodies of water. Among the spheroidal masses, a considerable amount of interstitial space had been left. Moreover, the crusts were much shattered, and frequently the interiors of the masses were penetrated by a multitude of cracks produced by shrinkage in cooling. Where the openings were of sufficient size to permit the free passage of superheated aqueous vapors from the water-impregnated sediments beneath, these gases appear to have rushed upward with great force and velocity, carrying with them quantities of finely comminuted dust from the lake beds and depositing it in the various interstices, in the form of a reddish-brown powder. Much of this is probably of a clayey nature, but it is so fine that it is not resolvable under the microscope, though some of it shows also grains of quartz and feldspar such as are found in the coarser sediments. Mingled with this material, there are many fragments of basaltic glass, whose nature is easily recognized. In some cases, these deposits appear to have filled open spaces of considerable size and in others to have penetrated minute cracks. At a quarry formerly worked near Great Notch, the igneous rock carries between the boulder-forms irregular blocks of this character, ranging in size up to a foot or more in diameter, at first sight counterfeiting inclusions of fine-grained sandstone. Examination under the microscope, however, shows the presence in this pseudo-sandstone of a notable admixture of glass, and the manner in which the material ramifies into cracks of the basalt suggests its origin. Hand-specimens may sometimes be found, in which the peculiar relation appears of small dike-like bands of reddish sandstone cutting the basalt. This reversal of the usual relations is interesting from its unusual character.

The formation of such deposits appears in some instances to have sealed the larger spaces left open after the consolidation of the igneous masses, but nevertheless the brecciated material around the boulders and the multitude of shrinkage-cracks often present would offer spaces of capillary size or larger and would undoubtedly render the piled-up masses of boulder-like structure above the lake beds distinctly more porous and susceptible to the circulation of water than the dense, massive basalt beyond the borders of the lakes. Field observations lead to the conclusion that such is clearly the case.

SOURCE OF THE WATERS WHICH EFFECTED RECRYSTALLIZATION AND OF
THE NEW MINERALS DEPOSITED

The query arises as to the origin of the circulating waters, whether meteoric or magmatic. The writer adheres strongly to the views enunciated by J. F. Kemp and other authorities that in the majority of metaliferous deposits the metallic minerals are almost wholly, and the solutions largely, of magmatic derivation. In the case of these zeolitic deposits, however, several lines of evidence lead to the conclusion that the waters were of extraneous origin. Field observation shows that the secondary alteration extends to within a few feet of the lower surface of the trap sheet, and points to the uprise of waters from the underlying shales. In its theoretical aspects, the situation is distinctly different from that of mineral veins. In the latter case, it is conceived that underlying masses of fused material, slowly cooling and crystallizing, give off various emanations which are forced to rise to the surface either through channels in the overlying rock, or through fractures in previously consolidated portions of the same mass, toward which the emanations are driven in their effort to find vent.

In surface flows, however, there is little obstruction to the escape of the vapors into the open air. It can hardly be conceived that the evolution of vapors from surface flows continues long after the complete consolidation of the magma, for the process of crystallization necessarily excludes the gaseous material. The structure of the Watchung sheet, in the situation which we are considering, indicates that consolidation was almost simultaneous with accumulation and, consequently, that the emanation of vapors must have ceased shortly after the termination of the flow. On this point, the following quotation from Professor Kemp's article on "The Role of the Igneous Rocks in the Formation of Veins"¹⁶ is pertinent.

The vapors contained in surface-flows of igneous rock pass off directly into the atmosphere, and therefore do no geologic work of this character. The most that could be expected of them would be small incrustations in the cracks in their upper and first chilled portions, such as the copper minerals and specular hematite found in the crevices of Vesuvian lavas.

The greater number of the secondary minerals are traceable to the primary minerals of the magma. Pectolite, prehnite, amphibole, the zeolites, hematite, chlorite etc., contain only elements which are present in feldspar, diopside and magnetite. There are others, however, such as datolite and various metallic sulphides, containing elements that are only

¹⁶ Trans. Am. Inst. Min. Eng. vol. 31 pp. 169-198, 1901

found in minute quantities at most in the normal basalt. These could not have been formed by a simple rearrangement of the ingredients of the primary minerals in contact with the circulating waters.

If the conditions under which the lava consolidated have been correctly interpreted, it is not difficult to account for the relative concentration of these latter ingredients. The elements which are in excess are those which have often been observed as sublimates deposited in crevices in lava flows. They differ from aqueous vapors in that the temperature of sublimation is high, and a relatively slight cooling causes deposition in the solid form, often near the point of derivation.

The elimination of vapors of B_2O_3 from consolidating lavas and their deposition in crevices appear to be normal phenomena of volcanic activity. Deville and Leblanc in their observations at Vulcano¹⁷ found that gas from one of the craters, issuing at a temperature above the melting point of lead and emitting flames, deposited boric acid. At the "soffionis" of Tuscany, jets of steam carrying boric acid emerge from the ground. The condensable vapors from the fumaroles of Monte Cerboli contain boric acid, together with ammonia and hydrogen sulphide. Borates are also found in hot springs in numerous volcanic regions,—Northern California, the Yellowstone Park, the Cordilleras of Coquimbo, Argentina, Tibet, on the sea of Azov and in other localities. Borates appear, indeed, to be among the commonest volcanic sublimates.¹⁸

A. von Groddeck¹⁹ has reviewed the geological occurrence of boron minerals and expresses the conviction that the borosilicates (tourmaline, axinite, datolite, danburite) and water-free borates (rhodizite, jeremejevite etc.) with the exception of boracite, appear exclusively as autogenic forms in eruptive, archean and metamorphic rocks; and that in the whole series of fossil-bearing strata, borosilicates which have been formed without doubt in situ are wanting. By the latter, however, he doubtless did not intend to exclude the effects of contact metamorphism by means of vapors given off by an intruded eruptive, by which tourmaline is frequently formed. He emphasizes the fact that axinite, danburite and datolite are only crevice and druse minerals, and he considers that the chief field of distribution of axinite and datolite is notably in basic eruptive rocks, hornblende schists, chlorite schists and green schists, in which tourmaline almost not at all or only seldom appears.

It is interesting to note, however, that at the contact of the diabase sill which forms the Palisades along the Hudson, the arkose Newark shales

¹⁷ F. W. CLARKE: "Data of Geochemistry," Bull. 330, U. S. Geol. Surv. p. 214, 1908.

¹⁸ A. GEIKIE: "Textbook of Geology," London, p. 269, 1903.

¹⁹ Zeitschr. deutsch. geol. Gesell., vol. 39, p. 253, 1887.

have been metamorphosed in places to a tourmaline hornfels,²⁰ while in the sill itself fissures are lined with nests of datolite.

Metallic sulphides, pyrite and chalcopyrite especially, are often observable among the secondary minerals but are rarely seen in the unaltered basalt. Although present in but small amount, there was undoubtedly some concentration of sulphides in and near the channels of circulation. This may likewise be accounted for by the emanation of sulphur compounds and sublimates of copper and iron given off by the magma. Covellite (CuS) has been found on the lavas of Vesuvius,²¹ and atacamite ($\text{CuCl}_2 \cdot 3\text{Cu}(\text{OH})_2$) is also found in crevices in the Vesuvian lavas, as well as the chlorides of iron.²² Among the fumarolic deposits in the crater of Vulcano,²³ which are partly sublimates and partly secondary products, A. Bergeat gives compounds of cobalt, zinc, tin, lead and copper. Sulphides have also been found as sublimation products at Vesuvius, formed perhaps by the action of H_2S upon volatilized metallic chlorides.

The small amount of fluorine represented by apophyllite may also be referred to the magma. Fluorite sometimes appears on volcanic lavas.²⁴ It is possible, however, that the crusts of glass retained sufficient fluorine to supply the very moderate amounts required to account for the formation of apophyllite.

Such sublimates given off by the consolidating Watchung lava were probably deposited in crevices and interstitial spaces upon a slight fall of temperature. The places favorable for deposition were the very ones which would be easily reached later by circulating waters.

The aqueous contents of the lava would be completely expelled into the open air. The source of the waters which effected alteration must therefore be sought elsewhere. A theory of derivation from meteoric sources by a process of underground circulation appears to fulfill the requirements of the situation and to offer no obstacle. The preexistent topographic depressions in the areas occupied by lakes, although buried by the flow of lava, would nevertheless be reached by waters flowing in from the edge of the sheet through the porous sedimentary strata lying just beneath the impervious mass of normally crystallized igneous rock. Having reached such depressions, the hydrostatic head would be that due to the difference in elevation between the depressions and the point of ingress. The somewhat loose and porous structure of the accumulations of lava

²⁰ ANDRÉ and OSANN. *Verh. des naturhist. med. Ver. zu Heidelberg*, n. f., vol. 1, 1892.

²¹ E. S. DANA. "System of Mineralogy," 6th edition, p. 68, 1893.

²² *Ibid.*, p. 165.

²³ F. W. CLARKE. *Op. cit.* p. 220, ff.

²⁴ F. W. CLARKE. *Ibid.* p. 274.

above the preëxistent lakes being favorable for percolation, the waters would naturally tend to rise to the surface. A circumstance favorable to this result lay in the fact that the lava retained a store of heat long after consolidation. Immediately after the latter event the temperature was so high that apparently all water which gained access was vaporized, notwithstanding the impediments to escape caused by restrictions of the channel; but as the temperature fell, the water level in this arm of the system of circulation slowly rose.

The system of circulation thus conceived is analogous to that of artesian flows, only modified in this respect, that the effective head was due not so much to a difference in elevation between the points of ingress and exit, as to vaporization of the water in the upper portions of the channels of exit. It embodies the principle by which C. R. Van Hise has sought to explain the deposition of metallic minerals in fissure veins.²⁵ Exception has been taken to Van Hise's conception of the origin of metalliferous deposits, and in this case, where the operation of the principle seems almost demonstrable, the effects are distinctly different from what he has conceived that they should be. In justice, however, attention should be called to the fact that present exposures show only the lower portions of the return channels, and the possibility that higher levels would present more similarity to metalliferous veins is not necessarily excluded.

An alternative hypothesis to the one which has been offered should be considered, that is, the descent of meteoric waters direct from the surface through the channels in which alteration was effected. The form of this hypothesis which would consider cold surface waters as the agent may be dismissed almost at once; the results are entirely at variance with it. At the present time, such waters are effecting the solution of certain of the minerals, are decomposing others to pulverulent or clayey masses, are oxidizing the iron compounds to hydrated, rusty-looking material, and in short are giving to the whole the appearance which is characteristic of surface weathering. There still remains, however, the idea that the action may have taken place while the rocks were still highly heated. Two chief objections appear. The leaching which the rocks experienced while alteration was in progress, as shown by the great reduction in iron and magnesia contents, and the marked migration of other compounds implies a fairly rapid circulation. It is difficult to conceive of the means by which this could be secured under the hypothesis which we are now considering. Secondly, the nature of the secondary minerals shows conclusively that the process was not one of oxidation. Surface waters passing through

²⁵ Trans. Am. Inst. Min. Eng. vol. 30, pp. 27-177, 1900.

several miles of underground channels might easily be deprived of their dissolved oxygen in several ways, but waters descending directly from the surface would necessarily produce oxidizing effects.

CHEMISTRY OF THE PROCESS BY WHICH ALTERATION WAS EFFECTED

The principal rôle of the water seems to have been to form a medium in which recombination of the elements of the rock could be easily effected. The minerals arising from the crystallization of the magma were those which were in mutual equilibrium under the conditions of temperature, pressure and concentration prevailing at the time of consolidation. In the presence of heated water, they were, to a slight extent, dissolved and were thereby transferred to a solution in which entirely different conditions of temperature, pressure and concentration were present. As the chemical equilibrium is dependent upon these factors, new reactions followed, and it may be profitable to devote a short space to an inquiry into the mechanism of the process. According to modern conceptions of ionization, a mineral or salt which is soluble in only minute quantities in water undergoes in such a solution a change by which the molecular grouping which characterized the solid is broken up into simpler groups termed ions.²⁶ A familiar example of this is the dissociation of AgCl , by which an Ag and a Cl ion are formed, carrying electrical charges which are equal and of opposite sign. The quantity of un-ionized AgCl in the solution is extremely minute, but is not negligible. Addition of a second salt, such as NaCl , which has an ion in common, drives back the ionization of AgCl and causes some to be deposited from the solution.

Chemical reactions between such salts in solution are believed to be chiefly of an ionic nature. If in the formation of new compounds, the quantity produced to satisfy the conditions of equilibrium demanded by the nature of the reacting substances is in excess of the solubility, a portion will be deposited in the solid state. Such a reaction abstracts ions and permits a further dissociation of molecules. This in turn permits more of the original solid to pass into solution, and in this manner the process is continuous and will proceed until all the available material has been used up.

The process thus described enables us to form a mental picture of the mechanism of alteration but does not suggest any explanation of the

²⁶ There has lately arisen some doubt as to whether the theories of ionization which have been founded upon older conceptions of the molecule represent with exactitude actual conditions and processes. Whether this is true or not, the idea forms a practical working hypothesis, not out of harmony with main facts, and of value accordingly. There is no tendency to abandon it, until a better substitute can be found.

forces by which the mechanism is urged forward. A little further insight into this is permitted by the fact, which has been ascertained to be true in many instances and is believed from thermodynamic considerations to be a general law, that in any association of compounds, the stable forms are those for which, under the given conditions of temperature, the vapor pressures are a minimum. The vapor pressures of solids are generally so minute as to appear almost infinitesimal, but are not therefore negligible. Even without the intervention of a medium of solution, molecular rearrangements take place. This is well established for metallic alloys and sometimes occurs with natural minerals. In general, however, a solvent is requisite, and in its presence the compounds having higher vapor pressures pass more readily into solution and those with least vapor pressures crystallize out.

The principle is of immediate applicability in explaining the facility with which the glassy crusts of the basalt were attacked by the circulating waters. A glass, as is well known, is, from the standpoint of physical chemistry, a greatly under-cooled liquid, in which crystallization has been checked by the viscosity. Its metastable condition is expressed in the excess of its vapor pressure over that which the component minerals would possess in their crystalline phase. It should therefore pass into solution more readily than the normally crystalline basalt, and such seems undoubtedly to have been the case. When the structure of the deposits is examined in the field, it is found that the secondary minerals form nests and pockets in the angular spaces between adjacent boulder-forms and wrap around them in bands in the situation in which the glassy crusts were originally developed. This is in part due, no doubt, to the fact that channels of easiest circulation followed such features, but the whole effect cannot be ascribed to the latter cause, for cracks produced by shrinkage or by deformation and passing through the interior of the boulders do not exhibit an equal development of alteration products. Confirmatory evidence of selective alteration of glass is found in the slides. In a number of instances, crystalline crusts of later minerals show structures which seem best explainable as survivals of original forms in glass. Recrystallization seems to have been checked, however, when the normal basalt was reached, and at times its effects almost disappear within the area of the thin section.

Slide 53 affords an instructive illustration of several of these points, as is shown in Plate XI, fig. 5. The microscope shows crusts of recrystallized minerals composed of prehnite, green amphibole, specularite, secondary albite and garnet. Although recrystallization has produced large crystals, an indication of a former structure survives in certain curved

bands and circular markings, suggesting in form the spherulites and similar features found frequently in the glasses. The resemblance to the chloritic nodules of fig. 2 and other sketches is obvious, and their occurrence in the present case is ascribed to a persistence of the spherulitic structure during recrystallization, the effects being preserved by an insoluble dust of TiO_2 or MnO_2 . Next to these crusts lies a narrow band in which the basaltic texture appears, but in finely crystalline development, and this is believed to mark the transition from glass to normal basalt in the original rock. Beyond this the texture is that of ordinary basalt. Recrystallization has been complete in the glass, except for the insoluble inclusions. In the finely crystalline border and in the normal facies of the rock, it has made sufficient advance to change the labradorite and diopside to prehnite and albite for a short distance without much disturbance of the texture. A little further, and both the texture and mineralogic composition are almost unchanged. Similar phenomena, showing that alteration was checked when the crystalline portion of the basalt was reached, are seen in several slides.

The fact that circulation was comparatively rapid had important results in the nature of the secondary minerals. Under these conditions, rapidity of solution or time taken for a mineral to form a solution of a certain strength, considered apart from total solubility or quantity present in a saturated solution, affects the nature of the changes which take place. When water is at rest in the interstitial spaces in a rock, a mineral which dissolves slowly may nevertheless reach a comparatively large total solubility, but if the water moves along channels of circulation, the amount of a slowly soluble substance which is taken up may never reach the amount necessary to carry forward sufficiently those reactions by which new minerals of less solubility containing its elements are deposited, and whatever amount goes into solution is removed altogether.

In the original basalt, magnesia and the oxides of iron reach a large percentage, being principal constituents of the diopside and magnetite. In the later minerals, the proportion of iron and magnesia is insignificant. In places, large masses of zeolitic minerals and calcite are found from which all compounds containing iron and magnesia have been removed.

Another effect of the circulation of the waters is that the substances dissolved are distributed through the solution, and new products may crystallize at a distance from those from which their elements were derived. With standing, interstitial water diffusion of dissolved substances can only occur through the slow action of osmotic pressure. Consequently, under the latter circumstances, the new minerals are much more apt to show intimate association with the old and little tendency to mi-

grate. In the more open channels of these rocks, the circulation was naturally much more rapid than in the capillary spaces. Some difference in concentration of dissolved material thus arises, and to this cause in part is probably to be attributed a marked banding parallel with cracks, which appears in many slides (Plate XII, figs. 3 and 4).

It appears from what has preceded that the glassy crusts of the boulder-like forms of basalt, lying next to relatively open channels and being in a chemically metastable condition, were naturally the portions of the rock most attacked. Alteration also follows the shrinkage cracks which penetrate the normally crystallized interior of the boulders and produces veinlike bands of secondary minerals in a breccia of unaltered basalt. There is also evidence that deformative stresses acted upon the region within a comparatively short period of the extrusion of the basalt, for shear-planes often show alteration. On the whole, however, movements or mechanical disturbances of any nature appear to have been almost negligible during recrystallization.

EVIDENCES AS TO TIME INVOLVED

It has not been found possible to determine from field observations what depth the flow of lava reached. At the quarry at West Paterson which has been mentioned, a section shows a thickness of 60-70 feet from the underlying shale to the surface of the ground, throughout which the characteristic features of pahoe-hoe are developed. This merely indicates what the minimum thickness was at this point and does not afford information as to the amount removed by erosion. There are indications, however, that the entire thickness of the First Watchung sheet was not involved, for in several localities, it has been found that the phase of the basalt in which the boulder-like structure is prominent passes under normal massive basalt, which appears to be a later flow. If this inference is correct, the whole history of alteration must have occupied only a comparatively brief period, geologically considered; for a second flow of lava, it would seem, would spread an impervious seal over the entire area and terminate the process at whatever stage had been reached. From other considerations also, the history is believed to have been brief. A sheet of lava, even several hundred feet in thickness, subjected to the cooling effects of percolating waters, would not retain its store of heat for a period which would be considered long from a geologic standpoint.

DEGREE OF SUPERHEATING OF THE SOLUTIONS

Although a rapid circulation has been spoken of in referring to the movements of the waters, this term has been intended merely in a relative

sense. It is believed that the openings in the rock were in general only of capillary dimensions. Great obstacles were thus presented to the easy circulation of water in the lower part of the channels and to the free escape of steam in the upper part. The temperature of vaporization was thus increased and superheating permitted. The maximum temperature possible under the circumstances was that corresponding to the pressure to which the water was subjected due to the hydrostatic head. Before the temperature fell to this point, the water was probably converted into steam and escaped in this form, but it is not believed that any important recrystallization was effected, until the temperature reached the point at which the liquid form was retained.

On account of uncertainties as to the thickness of the lava sheet, no close estimate can be made as to the probable degree of superheating, but some approximation can be arrived at. If a thickness of 300 feet is assumed as a probable maximum, the pressure at the bottom of a column of water of this height would be 130 pounds per square inch, or, taking account of additional atmospheric pressure, 145 pounds per square inch. The corresponding temperature of vaporization is 180°C. , and this may be considered as a probable maximum for the temperature which the solutions attained. No great degree of accuracy is to be claimed for such an estimate, but it will serve to show the probable order of superheating, and it indicates that no approach to the critical temperature of water (364.3°C.) was attained. Nevertheless, the minerals formed at this stage were similar to those which are found in highly metamorphosed schists and gneisses.

From the maximum, the temperature gradually fell as the mass of rock cooled. There are indications that, to the last, the temperature was somewhat above average climatic temperature. Among the products in the last stages, actinolite and chalcopyrite appear to have been formed, and these would hardly be expected as products of cold solutions.

The general process was that of the slow but constant and uninterrupted cooling of a mass of igneous rock through which aqueous solutions were percolating, deriving their temperature from the inclosing rock and cooling as it cooled. Material for solution was contributed by the basalt and by the previously evolved sublimates, and reactions followed by which new minerals were crystallized out. With progressive fall of temperature, conditions of chemical equilibrium within the solutions were shifted, and new species were deposited. In these later changes, the material which participated was derived both from the first-deposited minerals, which had now become unstable, and from new supplies of igneous rock to which access was gained.

The constant removal of material in solution undoubtedly enlarged the channels of circulation and, in places, produced cavities of considerable size. The most perfect crystals are naturally found in vugs of this kind. It does not appear, however, that such openings were at all necessary for recrystallization, and in most cases they were not present. The leaching appears to have been somewhat selective, for iron and magnesia are greatly reduced in the later stages.

II. PETROGRAPHY OF THE SECONDARY MINERALS

METHODS OF STUDY OF MUTUAL RELATIONS OF MINERALS

All the material for petrographic work has been obtained from the region lying between Paterson and Montclair Heights. In many places throughout this district, natural exposures of surface rock show evidences of the features which have been ascribed to underlying lake beds; but weathering has produced its usual effects of solution and decomposition, and such exposures do not afford desirable material for investigation. In a number of places, however, quarries have been opened for the purpose of obtaining road material or railroad ballast, and the fresh rock thus brought to view offers all the desired opportunities for study. A quarry at West Paterson and two at Great Notch have furnished the bulk of the material, but a considerable amount has been obtained from the waste-dump of a tunnel driven through the mountain near Great Notch a number of years ago for a water-supply system.

Before beginning the microscopic study of the relations which the various minerals bore to each other, it had been observed from macroscopic specimens that there was probably a definite order of deposition, and the writer presented a tentative determination of the sequence in a previous paper,²¹ although stating that there were often great difficulties in coming to a decision. As the petrographic work progressed, it was found necessary to revise previous ideas along several lines. Certain criteria which had been followed in accordance with general conceptions were found in this instance to be unreliable guides. The manner in which bands of granular calcite frequently appear between unaltered basalt and other secondary minerals may be cited as an example. The inference would be that the calcite was of earlier formation and was the first to be deposited on the basalt, but in numerous instances, it has been found to be later, and the latter relation is believed to be the general one. Similar effects appear with other minerals. Not infrequently, partial

²¹ Jour. Geol., vol. 16, pp. 297-321, 1908.

replacement causes an earlier mineral to appear to rest upon a later, and often those minerals which have an acicular habit seem to have had the ability to penetrate without difficulty through earlier-formed minerals of a more massive nature. For these reasons, it is believed that macroscopic observation alone is unreliable. On the other hand, microscopic observations have certain limitations, and it has been found advantageous to employ both methods as supplementary to each other.

In some respects, the zeolites are difficult subjects for petrographic study in random rock sections, when it is desired to differentiate the various members of the group, because of the rather negative character of the optical properties and the resemblances among different species. By careful study of known sections, however, it was found that differences of habit could be perceived which were often sufficiently distinctive for rapid identification, when checked in doubtful cases by further tests.

Throughout the various portions of the district covered, the relations of basalt and secondary minerals appear to be almost identical. There are differences in the same quarry which are as great as in parts of the field several miles apart. Such differences embrace the greater or less prevalence of the boulder-structure in places and the variations in the predominant minerals. The latter is probably an effect of the somewhat variable amount of sublimation products condensed in the crevices and of the facility with which solutions percolated through the channels.

LIST OF MINERALS

In the hand specimens the following minerals are observed to occur in quantity: quartz, datolite, prehnite, pectolite, analcite, chabazite, heulandite, stilbite, natrolite, laumontite, apophyllite, thaumasite and calcite. Gypsum is found, but is much more rare. Pyrite and chalcopyrite are frequent, but almost always in small grains. Galena and sphalerite are known to occur, but they have not been met by the writer. Hematite is often noticeable as a finely divided purplish dust disseminated through crystals of silicates and calcite. Tabular hematite crystals of larger size also occur. Gmelinite has been reported, but in a number of instances which have come under the writer's observation the specimens appeared to be more probably chabazite. The two, however, doubtless form a continuous series analogous to the plagioclases, and considerable variability in composition is to be expected. The relation of chabazite and of gmelinite to other zeolites probably presents no essential difference, and no attempt was made to distinguish the two in microscopic work. Scolecite has also been reported, but the writer has not met with satisfactory ex-

amples in hand-specimens. In several thin sections, small remnants of a mineral which is probably scolecite were observed in association with natrolite. The chemical relationship between natrolite and scolecite is somewhat similar to that between chabazite and gmelinite, but the group appears to present an example of a different type of solid solutions, namely, that in which the two components do not form a continuous series of mixed crystals, mutual solubility being limited and the end members being of distinct crystallographic forms.²⁸ Most of the specimens are plainly natrolite. Chlorites, probably of several species, are very abundant, but are more prominent under the microscope than in larger specimens. Serpentine is also found in thin sections, but is not of so frequent occurrence as chlorite. Several minerals occur in small quantities and have been identified by optical methods. Albite, garnet and several varieties of green amphibole were thus recognized, and their occurrence will be described in its appropriate place.

APPLICATION OF THE PHASE RULE

In reviewing the list of minerals given, it will be observed that the majority of the species are composed of those elements which form the plagioclase feldspars. Frequently there is an addition of the water molecule, but otherwise there is no change except in the relative proportions of the various oxides. Different forms of combination of soda (Na_2O), lime (CaO), alumina (Al_2O_3), silica (SiO_2) and water (H_2O) are accountable for all of the following minerals: labradorite, albite, quartz, prehnite, pectolite, analcite, chabazite, heulandite, stilbite, natrolite, scolecite and laumontite. In what follows, it is proposed to make a special investigation of the mutual relations of these species, based upon those methods of physico-chemical analysis which are embraced under Gibbs's phase rule. It is desirable, therefore, to make a preliminary inquiry into their composition.

The plagioclase feldspars: The recent investigations of A. L. Day and E. T. Allen²⁹ into the nature of the plagioclases, carried out in the Geophysical Laboratory at Washington, have shown almost beyond question that the albite and anorthite molecules form a continuous series of solid solutions of the type in which mutual solubility is unlimited and in which the fusing points of intermediate members lie between those of the isomorphous end-compounds. All the members from albite, $\text{Na}_2\text{OAl}_2\text{O}_3\text{-}$

²⁸ For analogues among artificial salts, see ALEXANDER FINDLAY: "The Phase Rule and its Applications," New York, p. 180, 1906.

²⁹ Am. Jour. Sci., 4th ser., vol. 29, pp. 52-70, 1909.

6SiO_2 , to anorthite, $\text{CaOAl}_2\text{O}_32\text{SiO}_2$, form, therefore, a single chemical phase. While natural plagioclases are, in the great majority of instances, combinations of albite and anorthite, there are reasons for thinking that soda anorthite ($\text{Na}_2\text{OAl}_2\text{O}_32\text{SiO}_2$) and lime albite ($\text{CaOAl}_2\text{O}_36\text{SiO}_2$) are also possible compounds. Doelter showed a number of years ago that fusion and slow recrystallization of some of the zeolites resulted in the formation of the compound $\text{CaOAl}_2\text{O}_36\text{SiO}_2$, and H. S. Washington and F. E. Wright³⁰ have lately investigated the composition of a feldspar from Linosa, whose formula from analysis corresponds to $(\frac{1}{3}\text{Ca}, \frac{2}{3}\text{Na})\text{Al}_2\text{Si}_3\text{O}_{10}$. They consider the most probable explanation of composition, on chemical and crystallographic grounds, to be the presence in solid solution of Ab, Or, An and a new compound, $\text{Na}_2\text{OAl}_2\text{O}_32\text{SiO}_2$, for which they propose the name carnegieite. They point out the resemblance of the mixed crystals which compose the feldspar to an unhydrated natrolite or mesolite. The matter has an important bearing upon the relations of the zeolites to the feldspars.

Quartz (SiO_2), *prehnite* ($\text{H}_2\text{Ca}_2\text{Al}_2\text{Si}_3\text{O}_{12}$ or $2\text{CaOAl}_2\text{O}_33\text{SiO}_2\text{H}_2\text{O}$) and *pectolite* ($\text{HNaCa}_2(\text{SiO}_3)_3$ or $\text{Na}_2\text{O}4\text{CaO}6\text{SiO}_2\text{H}_2\text{O}$) form each a phase of invariable composition. They are composed of the same oxides as the other members of the group which we are considering, but they do not present the same analogies in chemical structure.

The zeolites proper are all feldspathoid compounds whose resemblance in composition to the plagioclases has often been commented upon. The ratio of lime plus soda to alumina is 1 : 1, but the amounts of silica and water vary. They and the feldspars might all be embraced under a general formula $\text{ROAl}_2\text{O}_3\text{mSiO}_2\text{nH}_2\text{O}$. With certain of the species, the composition appears to be fixed, as in laumontite ($\text{CaOAl}_2\text{O}_34\text{SiO}_24\text{H}_2\text{O}$), but the majority of species form groups whose composition varies within rather wide limits. A typical example of this is the chabazite group, whose variability appears only explicable on the assumption that four distinct feldspar-like molecules enter independently ($\text{CaOAl}_2\text{O}_32\text{SiO}_24\text{H}_2\text{O}$), ($\text{Na}_2\text{OAl}_2\text{O}_32\text{SiO}_24\text{H}_2\text{O}$), ($\text{CaOAl}_2\text{O}_36\text{SiO}_28\text{H}_2\text{O}$) and $\text{Na}_2\text{OAl}_2\text{O}_36\text{SiO}_28\text{H}_2\text{O}$). The resemblance in composition which the various molecules bear to albite and anorthite and the assumed calcium-albite and sodium-anorthite is evident, and, like the ordinary plagioclase molecules, they appear to possess unlimited mutual solubility and form but a single phase. The structure of stilbite is similar, but less complex. The essential molecule appears to be $\text{CaOAl}_2\text{O}_36\text{SiO}_26\text{H}_2\text{O}$, with which is united in small but variable proportions the corresponding soda molecule,

³⁰ Am. Jour. Sci., 4th ser., vol. 29, pp. 52-70, 1909.

$\text{Na}_2\text{OAl}_2\text{O}_3\cdot 6\text{SiO}_2\cdot 6\text{H}_2\text{O}$. In heulandite we have similar relations, but with $5\text{H}_2\text{O}$. Typical analcite is $\text{Na}_2\text{OAl}_2\text{O}_3\cdot 4\text{SiO}_2\cdot 2\text{H}_2\text{O}$, but a small amount of lime usually enters. Similarly, natrolite is $\text{Na}_2\text{OAl}_2\text{O}_3\cdot 3\text{SiO}_2\cdot 2\text{H}_2\text{O}$, with a little lime sometimes present.

With most of the species, a little potash is frequently present and doubtless would be found by analysis in the Watchung minerals, but no distinctively potash zeolite has been noted.

From the manner in which the ratios of the molecules vary in the same mineral, it appears necessary to consider the form of union in the zeolites to be that of solid solutions rather than that which is usually termed chemical combination.

Each of the species enumerated may be considered a separate and distinct chemical phase. Including prehnite, pectolite and quartz, the phases present are as follows:

- | | |
|----------------------------|-----------------|
| 1. Labradorite and albite, | 7. Heulandite, |
| 2. Quartz, | 8. Stilbite, |
| 3. Prehnite, | 9. Natrolite, |
| 4. Pectolite, | 10. Scolecite, |
| 5. Analcite, | 11. Laumontite. |
| 6. Chabazite, | |

The phase rule states the number of phases which may be present in a system in equilibrium under given conditions of temperature, pressure and concentration.⁵¹

In the fundamental equation $P = C + 2 - F$,

P = number of phases,

C = number of components,

F = degrees of freedom (i. e., the numerical value of the variables, temperature, pressure and concentration).

In the system which we are considering, the components are five in number as follows: Na_2O , CaO , Al_2O_3 , SiO_2 , H_2O .

With regard to the numerical value which should be ascribed to F , some consideration is necessary. Temperature is without doubt in this case an independent variable. Pressure should be considered as dependent upon temperature, for in those portions of the system in which it was somewhat independent of temperature, no gaseous phase was present. Concentration probably varied from an independent function in the earlier stages to a dependent function in the later on account of the disturbing feature introduced by the presence of boric acid. After this had

⁵¹ ALEXANDER FINDLAY: *Op. cit.* p. 16.

W. D. BANCROFT: "The Phase Rule," Ithaca, N. Y., pp. 15, 1897

been taken completely into solution and had reacted with lime and silica to form datolite, no further supply was available, and it became an almost negligible factor. It appears probable from observations as to the period of stability of datolite which will be noted later, that from the beginning of alteration up to the time of formation of zeolites (a period in which garnet, amphibole, prehnite and pectolite were deposited, but in which feldspathoid compounds are small in amount or entirely lacking), concentration should be considered an independent variable, but in the later periods, in which zeolites are prominent, it should be considered to be nearly or altogether a function of temperature. To a minor degree, the presence of ferromagnesian silicates acted in a manner similar to boric acid in giving to the factor concentration an independent character, for the presence in solution of any compound having an ion in common with the feldspathoid series would affect their solubility, independently of temperature. The chief effect of these also is believed to have been in the first stages of alteration and to have become more nearly negligible during the later stages. The numerical value of F may be considered to vary from 2 in earlier stages to 1 in later.

Solving the equation for P , it appears that during the earlier period the greatest possible number of phases which could exist in equilibrium was five. The liquid and gaseous phases may be neglected, leaving three solid phases. During the later period, it seems at first that four solid phases in equilibrium would be a possibility, but a number of limiting conditions may be applied.³²

In all the feldspathoid combinations, which alone are now to be considered, the proportion $\text{Na}_2\text{O} + \text{CaO} : \text{Al}_2\text{O}_3 :: 1 : 1$ obtains. This decreases by one the number of phases possible and limits it to three. It appears also that with certain members of the series, there is merely a difference in the ratio of SiO_2 or H_2O to the other components. With analcite and natrolite, for instance, the only essential difference appears to be in the ratio of SiO_2 : analcite $= \text{Na}_2\text{OAl}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O}$; natrolite $= \text{Na}_2\text{OAl}_2\text{O}_3 \cdot 3\text{SiO}_2 \cdot 2\text{H}_2\text{O}$. These two species, therefore, cannot coexist in equilibrium under any change of conditions.³³ With heulandite and stilbite, the essential factor of difference appears to be the ratio of combined water.

With chabazite, the constitution is very complex, and it is doubtful what molecules could be removed without causing the crystals to break up. It is apparent, however, that one of the molecules cannot coexist

³² W. D. BANCROFT: *Op. cit.*, p. 234.

³³ In the application of the phase rule, there is no concern with the absolute composition of the molecule. Only the ratios are of importance.

with the anorthite molecule of plagioclase or with the essential molecule of laumontite and a second with the albite molecule, except at transition points, which, in the present case, are of little importance.

It is possible that there are other limiting conditions which are more obscure or regarding which present knowledge is insufficient. The evident relations, however, make clear the fact that not more than three of the feldspathoid minerals could exist together in stable equilibrium, and with several combinations not more than two could coexist.

It is necessary, however, to keep in mind the fact that, even under unstable conditions, solution may be so slow that portions of the mineral or even perfect crystals may persist through a prolonged period. There are indications that certain minerals formed during the first stages of recrystallization disappeared utterly and their nature can only be guessed at, but with the great majority of species, portions have survived throughout.

In applying the phase rule to the problem, the author does not wish to imply that he believes that a case of this kind is capable of treatment with a high degree of mathematical rigidity. The nature of the case renders this impossible. Each of the minerals present in addition to the feldspathoid compounds introduces an element of uncertainty which cannot be taken into account. It is believed, however, that these effects were rather feeble except with boric acid, which has been considered, and that, using some broadness of interpretation, the deductions of the phase rule are applicable.

The general results which should be expected are that the minerals formed from various combinations of soda, lime, alumina, silica and water should present a series in which one mineral after another will be found to have been deposited, to have survived through a certain period of stability, and then, with progressively changing conditions, to have passed again into solution, to be redeposited in new combinations. Regarding the order in which various species should appear, the phase rule affords no information. The tracing out of the sequence will form a chief feature of the petrographic portion of the present paper.

Without enlarging here upon the evidence which will be presented, it may be stated that the general inferences and conclusions drawn from the application of the phase rule appear to be fully warranted. Careful examination of hand specimens gives additional confirmation. Numerous examples have been collected which afford proof of the instability and breaking down and even the total removal of minerals. Such proofs are the etched and rounded surfaces of crystals, the development of porosity and drusy openings and finally the "negative pseudomorphs," or cavities

from which crystals have been removed. Before a detailed study was made, such effects were ascribed to ordinary processes of weathering, but they occur repeatedly in material in which oxidation and other normal effects of weathering are lacking, and it is frequently seen that later minerals have been deposited in the cavities formed by the removal of earlier.

The ferromagnesian minerals present a series parallel to the feldspathoid compounds, but in these rocks their quantity is too small to form the basis of an investigation similar to that which has been applied to the others. They present a number of interesting features, however, which will be described.

The phase rule supplies no information as to sequence of deposition, but simply shows how many phases can coexist in equilibrium. Nevertheless, if conditions were the same throughout the region, a sequence which has been determined in any instance should apply to all cases. In general, this appears to be true, but occasionally exceptions occur, implying local variations in concentration. This should be expected in a system of circulation of the kind described, depending probably upon the restrictions or freedom of flow and upon the tendency of minerals toward segregation.

Effects of this kind are exhibited especially by the ferromagnesian compounds. It is found, for instance, that the deposition of pleochroic green amphibole was conditioned largely by questions of concentration; and that, depending probably upon circulation becoming more free, amphibole which had been deposited was dissolved again at an early period, while under other circumstances its deposition was still continuing.

Another example is furnished by certain observations on analcite. From the prevailing evidence, it appears that in general analcite was one of the first of the zeolites to form, and, in several slides, it is found that chabazite and heulandite encroach upon it. On the other hand, a specimen has been found in which crystals of analcite rest upon heulandite in such a manner as to seem of necessity to imply later deposition. In this case, some local enrichment of the solutions in soda or impoverishment in lime is suggested. In later pages some inquiry will be made as to how far the sequence of minerals which has been observed should be considered an invariable one.

The processes of alteration may be divided along the lines indicated into two periods, which with their characteristic minerals are as follows:

Period I. Boric acid period.

Stage 1. Albite, quartz, garnet, amphiboles, specularite, sulphides.

Stage 2. Datolite, prehnite, pectolite, amphiboles, specularite, sulphides.

Period II. Zeolite period.

Analcite, chabazite, heulandite, stilbite, natrolite, scolecite, laumontite, apophyllite, amphiboles, chlorite, specularite, sulphides.

To these may be added a third period, whose significance will be shown later.

Period III. Calcite period.

Thaumasite, calcite, gypsum, amphiboles, chlorite, specularite, sulphides.

FIRST PERIOD OF ALTERATION

In the primary stage of alteration, temperature and pressure were at a maximum. The resulting minerals are those which are characteristic of a rather intense form of hydrothermal metamorphism. They include albite, arfvedsonite and other amphiboles, specularite (specular hematite), sulphides, garnet and quartz.

Albite

The feldspar of the unaltered basalt is a medium labradorite of about the composition albite 40, anorthite 60, occurring in hypidiomorphic crystals of lathlike habit, whose largest dimensions seldom exceed 0.5 mm. Among the alteration products, this is replaced by albite, whose size seldom falls below the maximum of the labradorite and ranges from this to a length of 3 mm. or more.

The process by which the transformation of labradorite into albite has been effected appears most explicable, if the plagioclase feldspars are regarded as members of a continuous series of solid solutions formed of the two molecules albite and anorthite, as appears to have been rendered certain by the researches of A. L. Day and E. T. Allen, to which reference has been made. The ratio in which the two will enter a crystal is dependent upon the composition of the solution in which crystallization is proceeding, and the crystal will continue in equilibrium with the solution only so long as the ratio of albite and anorthite molecules in the latter undergoes no change.

It is evident that, as the two molecules are chemically distinct, their fields of stability will not coincide and that, under a change of conditions, the crystals may find themselves in contact with a solution in which the anorthite molecule is unstable, while albite is not affected. Under these circumstances, the small amount of albite which dissolves in the liquid

soon effects saturation, but the anorthite enters reactions by which new compounds are formed from it and its identity is destroyed. The original crystals are thus placed in contact with a solution saturated with albite but lacking anorthite, and with such a solution only pure albite is in equilibrium.

At the beginning of the recrystallization of the basalt induced by the passage of heated aqueous solutions, the anorthite molecule appears to have undergone reactions by which garnet and datolite and possibly other compounds were produced from it. Albite under the new conditions was stable. Anorthite was therefore continually leached out of the labradorite, while albite was deposited in its place, a chief feature of the process being probably a growth of crystals at the expense of others in the vicinity.

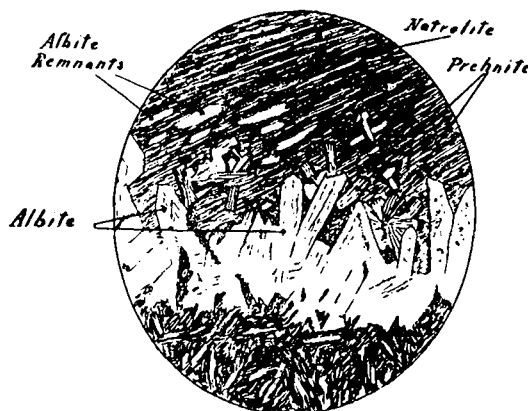


FIG. 3. Basalt bordered by vein of albite, prehnite and natrolite. Albite is developed from the labradorite of the basalt. Crystals of prehnite rest upon it and both are replaced by natrolite. $\times 35$. Slide 127.

In figs. 3-7 and in Plate XI, figs. 2, 3, albite is shown in characteristic forms and in typical relations with the basalt and with secondary minerals. Slide 127, a portion of which is sketched in fig. 3, was taken from a hand specimen in which the ordinary dense basalt is seen to be cut by narrow veins of secondary minerals. Under the microscope, the basalt presents the usual microcrystalline development, and the veins are found to consist of natrolite, prehnite and plagioclase feldspar. The crystals of the last occur in interlocking aggregates of varying crystallographic orientation. They form borders to the basalt and project at various angles into the veins, whose principal filling is natrolite. Crystals 0.5 mm. in length are not uncommon. Most of the crystals show albite twin-

ning repeated in two or three stripes. Terminal faces are often well developed.

These features characterize the secondary plagioclase in most of the slides in which it occurs. The identification of the plagioclase as albite has been determined by numerous tests. The statistical method of Michel-Lévy, applied to a great number of crystals, gives maximum extinction angles of 15° to 16° . The index of refraction is slightly less than that of balsam; optical character, positive. Sections perpendicular to the acute bisectrix Z give extinction angles of $13\frac{1}{2}^{\circ}$ to 15° , measured from the 001 cleavage. These tests are mutually confirmatory and indicate a practically pure albite.

An interesting feature observed in many instances is that the crystals of vein feldspar are plainly in crystallographic continuity with the plagioclase laths in the walls. It is notable, however, that among those which project farthest, the larger and stouter forms show a tendency to parallelism in a direction at right angles to the walls, as if their growth had been favored at the expense of those which projected at more acute angles. The average of size is decidedly larger than that of the plagioclase in the unaltered basalt. It is what might be expected if the original feldspars had grown until they occupied the space left by removal of diopside and magnetite, and in some cases, the relations are such as to force the conviction that this has occurred.

In the process of chemical readjustment by which crystals of a solid solution place themselves in equilibrium with the surrounding medium, when the composition of the latter becomes altered, two methods seem possible. The crystal might wholly dissolve and new and distinct crystals of the stable form might be deposited, or the identity of the crystal might be preserved, while the excess of one constituent was removed by solution and its place taken by a sufficient amount of the second constituent to supply the deficiency. The evidence of the thin sections described indicates that the process was to a large degree at least of the latter nature, the identity of the crystals being preserved. It is probable that this method of substitution, however, can advance only when the liquid in contact is very mobile and capable of penetrating within the interior of the crystals. If of the viscous constituency of most magmas, the contact would be merely at the surface and exchange between solid and liquid could proceed only through the almost infinitely slow agency of osmotic diffusion in the solid.

Albite in much larger crystals and showing a very different form of development appears in several slides. In these, the process of readjustment by which labradorite was transformed into albite appears less evi-

dent than is the case with the smaller form of albite crystals, but it is believed that the same process prevailed in a slightly modified form. Crystals of the larger type are best seen in slides 133 and 134. The hand

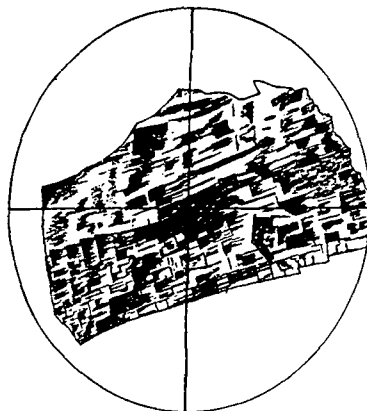


FIG. 4. Single crystal of secondary albite showing both carlsbad and albite twinning. Crossed nicols. $\times 35$. Slide 133.

specimen from which both of these slides were prepared shows numerous clear, glassy crystals imbedded in pectolite. The microscopic appearance

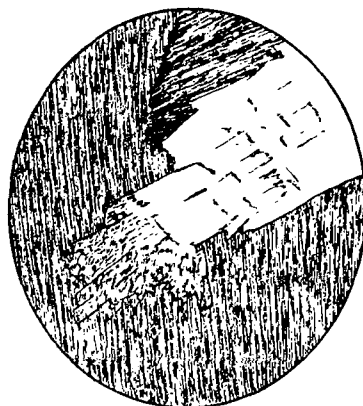


FIG. 5. Crystal of secondary albite partly replaced by pectolite. Outline is corroded and interior clouded in places, and pectolite fibres are advancing across it. $\times 35$. Slide 134.

of a single crystal between crossed nicols is shown in fig. 4, and one in which the albite has been partly replaced by pectolite is shown in fig. 5.

The appearance is so unlike that of the commoner form that numerous tests were made to establish its identity. Some of these were as follows:

1. All sections giving maximum interference (perpendicular to optic normal) give practically parallel extinction.
2. Sections perpendicular to both albite twinning and to basal cleavage give extinction angles of 13° to 16° .
3. Crystals which were found to be nearly or quite perpendicular to the acute bisectrix Z give extinction angles of 11° to 18° with the basal cleavage.
4. In most crystals, a combination of albite and Carlsbad twinning is found. By selecting those crystals which give approximately maximum extinction for albite lamellæ in both halves of the Carlsbad twin, corresponding angles of extinction were found as follows: $12\frac{1}{2}^{\circ}$ and $13\frac{1}{2}^{\circ}$, $12\frac{1}{2}^{\circ}$ and $13\frac{1}{2}^{\circ}$, $13\frac{1}{2}^{\circ}$ and $14\frac{1}{2}^{\circ}$, $11\frac{1}{2}^{\circ}$ and $13\frac{1}{2}^{\circ}$.
5. The index of refraction is less than that of balsam.

With all these facts in mutual accord, it can hardly be doubted that the mineral is nearly or quite pure albite.

The crystals are many times the size of the form of albite first described, attaining a maximum length of 3 mm. or more, being thus comparable with the crystals in granitoid rocks. Carlsbad twinning is very common. The two parts are joined along a somewhat irregular line, as shown in fig. 4. The most characteristic feature is the curiously mottled appearance, due to the irregular manner in which the albite twinning lamellæ are placed in juxtaposition. The lamellæ do not in general traverse the whole length of the crystal, but often they stop abruptly. Between crossed nicols, the different interference colors thus brought to view in a single crystal have a patchwork effect, and careful study shows that, curiously enough, the form of the patches outlined is suggestive of a multitude of lathlike plagioclase crystals disposed more or less at random. It is conceived as possible that this is a clue to the manner in which these large crystals of albite originated; that in fact each individual is built up of a multitude of labradorite laths of the original basalt, which have been incorporated within a single expanding crystal, and whose molecules have been swung into approximately correct crystallographic orientation. That the crystallizing forces of the major crystal were not able to exert rigid control throughout is indicated by certain anomalies in extinction and by a curving of the ends of the crystals which is frequently shown.

In slide 129, several groups of albite crystals of the large type shown in slides 133 and 134 appear in the midst of apophyllite. The borders are corroded and the interiors muddy, and they are plainly remnants giving way to the apophyllite. It is not improbable that a great deal of secondary albite of this form appeared among the first products of altera-

tion, but little of it now survives. Intermediate forms between the large and small types described appear in several slides, for instance, 68, 70, 119 and 120, generally in isolated crystals or small groups, and show active replacement.

A frequent mode of occurrence of albite in these rocks is as part of the filling of small, sharply defined veins which often cut the walls of quarries. They have a northeasterly and southwesterly strike. The veins bear evidence of being the result of deformative stresses which have affected the region. The presence of albite, which was probably one of the first products of recrystallization and which was formed while the rocks were still in a superheated condition, indicates probably that the regional adjustments by which the cracks were formed took place within a comparatively short period after the extrusion of the lava.

Replacement of Albite

A normal association of albite with fibrous amphibole, garnet and specularite, in which the relations are but little obscured by further stages of replacement, appears in slide 98 (Plate XIII, fig. 1). In this slide a normally crystalline basalt is crusted with a variety of secondary minerals, which in places show evidence in their structure of having replaced a glassy crust of lava. The primary alteration seems to have resulted in albite, green amphibole, garnet and specularite, which appear in close association next to the unaltered basalt. Further out, these products are giving way to prehnite, and at one place shown in the sketch, chabazite is the replacing mineral.

The typical manner in which replacement attacks the albite is shown in Plate XI, fig. 3, from slide 62. In this slide, finely crystalline basalt is cut by veinlets (probably originating as shrinkage-cracks in cooling) whose filling consists principally of albite and prehnite. In the more minute veins, where there is little prehnite, the albite looks perfectly fresh. The crusts, between crossed nicols, have the appearance shown in Plate XI, fig. 2, the albite being finely granular next to the basalt and assuming a development of larger crystals at a short distance. In the more open areas, where prehnite is abundant, the albite crystals have assumed a spongy appearance (suggestive of ice which has been exposed to the sun) and have lost their sharpness of outline. Fanlike groups of prehnite appear to spring up at any point within the massed crystals of albite and gradually encroach upon them, as shown in Plate XI, fig. 3. The albite in association then appears corroded at the edges and turbid within. By a continuation of the invasion, albite has disappeared over

large areas, and the only trace of it left is a hazy or muddy appearance of the prehnite. In many places, the patches of turbid material within the prehnite still show in their outlines the appearance of albite groups.

In slide 65, a similar process of replacement has produced the results shown in fig. 6. As in slide 62, veinlets of albite and prehnite are found cutting a crystalline basalt. The albite lies next to the walls and gives way rather abruptly to prehnite, but a few outlying crystals of albite, greatly corroded, appear to have survived within the prehnite at some distance from the walls. Still farther out, phantom forms of former albite crystals are outlined by a slight turbidity in the prehnite. They are completely replaced by prehnite and can be connected with albite only by the appearance of the terminal crystal faces. These, however, are characteristic. The radiating and fanlike groups of prehnite crystals

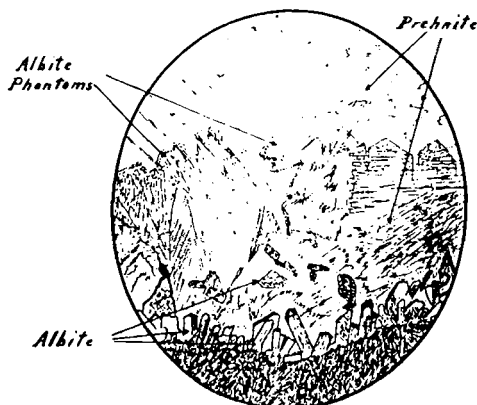


FIG. 6. Replacement of albite by prehnite. Phantom forms of former albite crystals are dimly perceptible. $\times 35$. Slide 65.

are superimposed upon the former structure of the albite, but in places the albite has had an influence upon the crystallization of the prehnite.

In slide 63, minute veins similar to those described are filled with albite and datolite. The albite crystals occur in interlocking aggregates of varying crystallographic orientation. In places, they occupy the veins to the exclusion of all other material, or they may project at various angles from the sides of the vein into masses of datolite. Crystals up to 0.5 mm. in length are not uncommon, and the general average of size is noticeably larger than the plagioclase rods in the walls. In most places, there is no evidence of corrosion of albite at contact with datolite, but in some areas, phenomena of replacement similar to those which have been described in association with prehnite appear.

Hand specimen 70 consists of a crystallized mass of secondary minerals, among which natrolite, heulandite and datolite are prominent. The slide shows at one point a group of rather large albite crystals within the datolite, evincing much corrosion of outline.

In 133 and 134, the hand specimen shows large, apparently clear crystals of albite, imbedded in beautifully crystallized pectolite. Under the microscope, the albite appears quite turbid in places, and the outlines show evident encroachment of pectolite fibres (fig. 5). In 136, albite and natrolite appear as the filling of a shear vein in dense trap. The manner in which natrolite is replacing albite is sketched in fig. 7. It is evident that the orientation of natrolite has been strongly influenced by the direction of elongation of the albite crystals which they are replacing. In 127

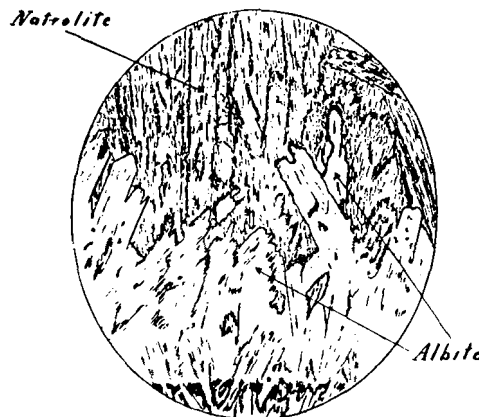


FIG. 7. Replacement of albite by natrolite. $\times 35$. Slide 136.

(fig. 3), the relations are similar, but in this case an intermediate stage is shown, for little groups of prehnite crystals are perched upon the partly corroded terminal faces of the albite. The prehnite, however, did not attain a strong development, and both it and albite are giving way to natrolite. It appears probable that, conditions being suitable for the formation of natrolite, albite would yield to its advance with great facility on account of the similarity in the composition of the two minerals (albite = $\text{Na}_2\text{O}, \text{Al}_2\text{O}_3, 6\text{SiO}_2$; natrolite = $\text{Na}_2\text{O}, \text{Al}_2\text{O}_3, 3\text{SiO}_2 + 2\text{H}_2\text{O}$).

In 136, numerous remnants of albite are buried in natrolite fibres, and the natrolite appears to have always assumed a position parallel to the elongation of the albite. Slides 56 and 137 resemble 136 as being minute veins in zones of vertical shearing, which produced sharply defined cracks in quarry walls. In 56, the filling of the veinlet appeared to the naked

eye to be wholly stilbite, but with the microscope it is found that this is replacing albite, of which numerous remnants are seen. Similarly in 137, small albite crystals are being replaced by chabazite. In 123, veinlets, which may be due either to shrinkage on cooling or to shear movements, contain albite, datolite and calcite, whose succession is in the order given. In 129, a large crystal of apophyllite carries several isolated inclusions of turbid-looking and much corroded albite of the larger type described.

Numerous other slides show more or less albite, in some places in rather fresh-looking crystals and in others in isolated groups of greatly clouded appearance. Occasionally its alteration has proceeded so far that the only trace of its former existence lies in turbid patches in other minerals, suggestive of albite merely by their form.

As to the mode of occurrence of albite in these rocks, it has been found most frequently in minute veins, but this may be due to the fact that under such circumstances the formation of the first products of alteration would tend to seal up the veins and render it difficult for later solutions to enter and attack the albite. In 95, the relations are different. Small, amygdaloidal cavities are filled with secondary products, among which albite and green amphibole and minute grains of garnet appear. It seems also that phenocrysts of diopside of the primary crystallization of the magma have been replaced by albite groups, illustrating two facts observable in many cases in these rocks, namely: that ferromagnesian minerals tend to disappear and that the replacing minerals often pay little attention to the composition of the material replaced. Slide 62 also shows this replacement of diopside by albite. Slide 100 illustrates again the filling of amygdaloidal cavities by albite and amphibole, but in this instance later stages of alteration are shown, and chabazite, calcite and chlorite are prominent.

In several slides, albite is found in a mode of occurrence which has been referred to before in discussing the manner in which solutions attacked the rock. The original texture of the normally crystalline basalt is preserved, but the mineralogic make-up is quite altered. In such cases, the laths of labradorite of the original have frequently been altered to albite. Slides 62 and 118 show this effect plainly.

The position of albite in the replacement series is determined by the minerals in respect to which it has been found to exhibit instability. Datolite, prehnite, pectolite, chabazite, stilbite, natrolite, apophyllite and calcite have all been found in relations which indicate energetic corrosion and replacement of albite, and these minerals are therefore regarded as belonging to later periods of alteration. This conclusion is confirmed by the relations of these minerals among themselves and to other min-

erals and may be regarded as established. On the other hand, albite has not been found in any case to replace minerals other than those of the primary consolidation of the magma. It is concluded, therefore, that it appeared among the first results of alteration. At the same time quartz, green amphibole, garnet, hematite and sulphide minerals (pyrite and chalcopyrite) appear to have been produced. Where albite is found in association with these, the contacts show no alteration on either hand.

In the breaking-down of the original labradorite into albite and various lime minerals, the presence of boric acid and other sublimates was undoubtedly a factor of great importance. Similar results of the action of hot aqueous solutions upon igneous rocks, by which an original lime-soda feldspar has disappeared and albite has been deposited in its place, are of fairly common and widespread occurrence, and it appears that in general the albite molecule possesses a much wider range of stability under conditions which are apt to occur in nature than does the anorthite molecule. While the more calcic feldspars normally occur as products of crystallization from igneous fusion, albite is most frequently found under conditions indicative of deposition from aqueous or aqueo-igneous solution at much more moderate temperatures. Fouqué³⁴ observed that albite never occurs individualized among the crystals of the volcanic rocks. Dana³⁵ mentions instances of its formation under metamorphic conditions or as a product of aqueo-igneous deposition as follows:

It is found in disseminated crystals in granular limestone, thus in the limestone (Jura and Trias) of the Col du Bonhomme, near Modane in Savoy; also in microscopic crystals with quartz and orthoclase in limestone at Meylan near Grenoble; in minute crystals in fossil radiolarians in limestone near Rovigno, Province of Pavia, Italy, also in the limestone itself; in limestone at Bedous, Basses Pyrénées, at the contact with diabase. Some of the most prominent European localities are in cavities and veins in the granite or granitoid rocks of the Swiss and Austrian Alps, associated with adularia, smoky quartz, chlorite, titanite, apatite and many rarer species.

An instance of the passage of a lime-soda feldspar into albite is cited by Dana³⁶:

Münzing has investigated the Pitschthal pericline and finds that the crystals consist essentially of an oligoclase, rich in soda, upon which albite has been deposited in parallel position, especially in the cavities of the original crystals.

W. C. BRÖGGER,³⁷ in following out the paragenesis of the minerals of

³⁴ C. HINTZE: "Handbuch der Mineralogie," Leipzig, p. 1433, 1892.

³⁵ "System of Mineralogy," 6th ed., p. 331.

³⁶ *Ibid.*, p. 1026.

³⁷ *Zeitschr. für Kryst. und Min.*, vol. 16, p. 167, 1890.

the nepheline-syenite pegmatite veins of southern Norway, notes a development of secondary albite in a form of microperthitic intergrowth with orthoclase along cleavage lines. He finds evidence that the formation of this secondary albite belongs to a period of mineral-formation previous to zeolite-deposition, and continues:

One finds also at times that the albite of the microperthite was later removed again during the zeolite-phase, whereby quite cellular, spongy, porous feldspar-remnants result. Perhaps a small part of the analcite of the vugs may even have been formed at the expense of the dissolved albite.

The process and results described by him appear to be closely analogous (taking account of somewhat different conditions) to those which occurred in the Watchung rocks.

W. LINDGREN,³⁸ in discussing the minerals characteristic of various zones of vein-formation, considers soda-lime feldspars unstable in all vein zones, while albite is found in contact-metamorphic deposits, in deeper vein zones and in middle vein zones. The physical conditions prevailing in the Watchung rocks during the short period of formation of albite appear to have corresponded, so far as can be inferred, to those of deeper or middle vein zones.

The stability of the albite molecule, under conditions under which anorthite breaks down into other compounds, gives rise at times to the special effect which Sir Archibald Geikie³⁹ terms "albitisation,"

a process in which, while the lime of the plagioclase is removed or crystallized as calcite, instead of forming a lime-silicate like epidote or zoisite, the rest of the original mineral recrystallizes as a finely granular aggregate or mosaic of clear grains of albite.

F. Becke⁴⁰ describes an occurrence of secondary albite which offers considerable similarity to the manner of occurrence in the Watchung rocks. His explanation of its formation is interesting and suggestive in this connection. Referring to changes in the rock after consolidation, epidote and zoisite are recognized among the products, and in the same category is placed albitic plagioclase, which, in the form of irregular veins of crystallographically parallel orientation, penetrates the original plagioclase crystals. He continues:

I cannot recognize in mechanical effects upon the rock the origin of these new forms. I would much rather believe that our rocks, after the magmatic consolidation was finished, were for a long time under other conditions, in

³⁸ "The Relation of Ore-Deposition to Physical Conditions," *Econ. Geol.*, vol. 2, pp. 105-127, 1907.

³⁹ "Textbook of Geology," London and New York, vol. 2, p. 790, 1903.

⁴⁰ "Petr. Stud. am Tonallit der Rieserferner," *Tsch. Mitth.*, vol. 13, p. 420, 1892-1893.

which the combinations created by consolidation did not present the condition of most stable equilibrium; especially does this apply to the basic plagioclases, whose silicates tended toward a breaking-up into soda-aluminum silicate (albite) on one hand, into lime-aluminum silicate (zoisite, epidote) on the other.

In this citation, Becke brings out clearly the conception that the stability of a mineral compound depends upon the conditions surrounding it, and that a compound which is stable under the conditions of its formation may become unstable under later conditions, and, if opportunity is afforded, will recrystallize in stable form. The writer wishes to emphasize this point on account of the manner in which a mineral is often spoken of as being very stable or very unstable. Unless the surrounding conditions are known, these terms have little meaning, for a reaction which proceeds in a certain direction under given conditions will (generally) proceed in the opposite direction under a reversal of conditions. This is very clearly expressed by C. R. Van Hise, who, in his work on "Metamorphism,"⁴¹ refers repeatedly to the manner in which reactions occurring in the zone of katamorphism are reversed in the zone of anamorphism, but many writers appear to ignore the principle. Throughout the present paper, one of the chief objects which has been kept in view is to show how quickly minerals respond to an alteration of external conditions, and express this facility of response in recrystallization, if opportunity is given.

Hintze⁴² gives numerous additional illustrations of the formation of secondary albite. In the vein-granite of the Riesengebirge, it is found in druses in the form of clustered rosettes or cockscomb crystal-aggregates. At Striegau, it covers walls of vugs and at times rests upon potash feldspars; in the Harz, it occurs in beautiful crystals in gabbro; near Marburg, Hesse, it forms a later deposit in crevices in diabase.

These various illustrations of the occurrence of secondary albite are cited to show its relations to the feldspars of the original rocks and are believed to confirm the view held by the writer that the various molecules which enter into feldspars are chemically independent and that the anorthite molecule (in some cases also the orthoclase molecule) may enter new combinations without affecting in any manner the stability of the albite molecule.

Quartz

Unlike albite, whose crystals are generally minute, quartz is often prominent in hand-specimens. The crystals frequently attain a thickness

⁴¹ Monograph 47, U. S. Geol. Surv., pp. 170, 172, 176, and especially 364-368.

⁴² *Op. cit.*, pp. 1433-1454.

of 10–15 mm., and much larger ones occasionally appear. It is generally colorless, but not infrequently amethystine, and more rarely smoky. The faces shown are usually simple combinations of prism and pyramid. In addition to the groups of euhedral crystals which are most common, granular aggregates sometimes occur, as well as deposits of chalcedonic silica.

Frequently specimens are found which show numerous cavities, indicating plainly the removal of groups of crystals of some mineral which was intergrown with the quartz. Most of the cavities are diamond-shaped in cross-section, but others are apparently rectangular. They are undoubtedly due to the removal of groups of crystals, but the forms do not suggest those of any of the species known from these localities. It appears most probable that among the various transformations experienced one or more minerals disappeared completely. There are indications that formation and removal occurred early in the history of the series, for at times the cavities have been filled with later-deposited quartz.

Replacement of Quartz

In slide 70, of which the hand specimen consists principally of datolite, heulandite and natrolite, the datolite, under the microscope, is seen to contain at one place a small group of albite crystals, greatly corroded

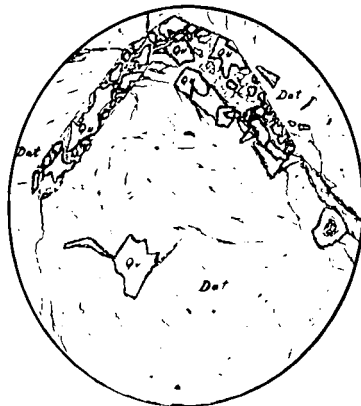


FIG. 8. Fragments of quartz outlining former crystal, now almost completely replaced by datolite. $\times 35$. Slide 70.

as previously described, and scattered through the datolite are minute remnants of quartz. One occurrence takes the form shown in fig. 8. It is clear that the scattered particles are portions of the pyramidal termina-

tion of a single quartz crystal, which has been almost completely replaced by datolite. There is little turbidity in either quartz or datolite. In

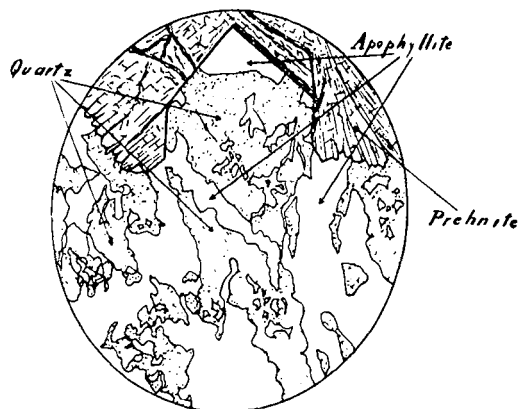


FIG. 9. Fragments of quartz with prehnite being replaced by apophyllite. Quartz is conventionally stippled to distinguish it from apophyllite. $\times 35$. Slide 138.

numerous other places, parting planes in the datolite appear to outline former quartz crystals, that is, the replacement of quartz has given rise

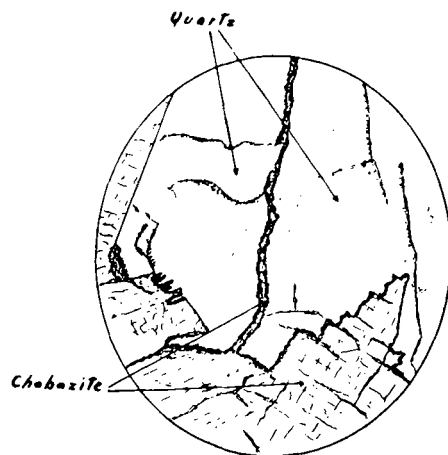


FIG. 10. Quartz cut by vein of chabazite and corroded by same on margin. $\times 35$. Slide 146.

to roughly pseudomorphic forms of datolite. In 125, also, pyramidal quartz crystals have been slightly replaced by datolite.

In 138, a large apophyllite crystal carries a great variety of inclusions, most of which are in process of replacement by the apophyllite. They comprise quartz, garnet, amphibole, prehnite, pectolite and calcite. Fragments of quartz are numerous. Relations to both apophyllite and prehnite are shown in fig. 9. The prehnite carries a sharply defined impression of a quartz pyramid, but only portions of the quartz crystal are left. The remainder of the space is occupied by apophyllite. The relations indicate that prehnite was deposited upon the terminal pyramid of a quartz crystal and that later both were removed during the formation of apophyllite or were partly incorporated within the latter.

In 87, fragments of quartz are inclosed in prehnite and pectolite in such a manner as to indicate replacement by both. In fig. 10, the rela-



FIG. 11. Replacement of quartz (clear) by chabazite (blocky). The quartz extinguishes nearly as a unit. Outside the field the radiating crystals of quartz spring from a centre. $\times 35$. Slide 77.

tions of quartz and chabazite in slide 146 are illustrated. The contact along one margin is perfectly straight, and it appears that corrosion has had no effect there. In other places, the quartz is greatly attacked, and chabazite has replaced it. A narrow vein of chabazite crosses the crystal, probably following some minute crevice which gave access to the solutions, and in various places lines of turbidity show the manner in which replacement begins.

In 77, replacement of quartz by chabazite in somewhat different relations appears. Heulandite also replaces the quartz. It is believed that the slide as a whole illustrates a frequent form of attack and replacement of quartz by later minerals. Figs. 11 and 12 show characteristic features. The original habit of the quartz in this slide seems to have been as groups

of radiating crystals, the individuals increasing in size outward. There was also probably a small amount of the unknown mineral whose removal by solution left the prismatic cavities previously described. In this case, the cavities have been refilled by heulandite, but are still sharply outlined. In other parts of the slide, quartz is giving way to heulandite, but in such cases the habit of the heulandite is different from that shown in its deposition in the open cavities.

In fig. 11, some indication of the radiating form of the quartz can be distinguished, and outside the field the center of radiation is plainly seen. The replacement by chabazite has taken a most irregular form, but the influence of the crystallographic character of the quartz upon its solution and removal are perceptible in places.

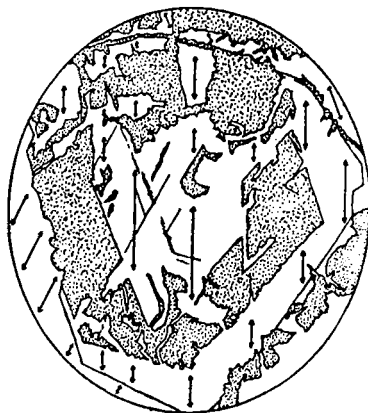


FIG. 12. Replacement of quartz (clear) by heulandite (stippled). Arrows show direction of vertical axes of quartz crystals. Many of the crystallographic lines approach parallelism, but few are strictly parallel. $\times 35$. Slide 77.

In fig. 12, the major portion of the field is occupied by a single quartz crystal, in nearly basal section, as is shown by the interference figure. Surrounding it are other quartz crystals of various optical orientation. Replacement by heulandite has taken the form shown, extremely irregular but yet plainly governed more or less by differences of solubility in the quartz.

Plate XI, fig. 4, illustrates one of the complex zeolitic groupings often found. Even at this stage of alteration, vestiges of the original texture of the rock and of the results of mechanical action upon it can be seen in many places. Phenocrysts of diopside are quite numerous outside of the field illustrated. Traces of plagioclase laths can be made out, and the subcircular chlorite nodules due originally to resorption of olivine are

plentiful. There are many fracture-lines, into some of which a reddish-brown deposit of sand and clay has been carried from the underlying lake beds. Most of the slide is in the stage of recrystallization in which zeolites are prominent, but in places has advanced to the stage of calcite and kaolin. Stilbite and analcite can be recognized, and others may be present. In the illustration, the radiating groups somewhat simulating forms of vegetation are probably crystals of quartz surviving from a much earlier period. They appear to branch from minute cracks. In some portions of the slide they show strongly the effects of solution. As regards the replacing mineral, it is not possible to be more specific than to say that it is probably a zeolite. In 125, quartz crystals which have begun

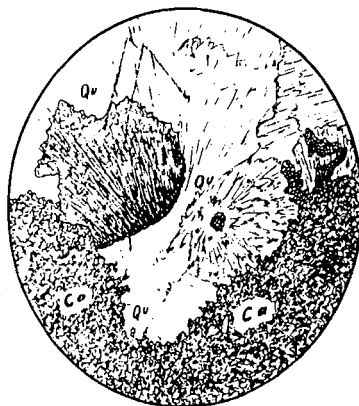


FIG. 13. Replacement of quartz by calcite. The quartz has the fan structure generally supposed to be typical of vein quartz, but it has probably replaced basalt. The granular-looking calcite shows features which are plainly a survival of those in the quartz. $\times 35$. Slide 58.

their growth in similar groups radiating from cracks have reached a large size.

In slide 58, a dense, normally crystalline basalt is crusted with quartz and chalcedony. The quartz has the fanlike crystallization supposed to be typical of vein quartz, although certain circular lines and markings (similar to those shown in fig. 2, though not so pronounced) suggest that much of the quartz may have replaced a glassy crust of basalt. The chalcedonic SiO_2 has a fibrous appearance and negative elongation. Within the basalt, the normal texture is preserved, but with crossed nicols it is seen that in places both diopside and plagioclase have been replaced by a mosaic of quartz. In the outer crusts, calcite in a finely granular form is working in and replacing the quartz. It is developed most

strongly in a band lying between the quartz and the basalt, and it might appear at first that it was earlier than the quartz, but its general character and the manner in which it branches minutely into the quartz are indicative of replacement. In addition, certain features shown in fig. 13 can be explained only in this way. Inclusions, plainly original in the quartz, survive within the replacing calcite.

This form of replacement, by which later calcite has worked in between crystallized quartz and basalt, seems to have been very common. Another good example is found in slide 41, and in repeated instances hand specimens are found to show a narrow band of calcite between the two, which, without microscopic examination, would naturally be inferred to be earlier than the quartz. A similar development of calcite as regards other minerals is referred to elsewhere.

It is not easy to understand why a deposition of calcite should effect the simultaneous removal of quartz or why the solution of quartz should cause the deposition of calcite, since the two minerals have no ion in common. It is known, however, that the solubility of a salt in water is often diminished by the presence of another salt, even when there is no common ion.

The observed relations indicate that quartz is earlier than datolite, prehnite, pectolite, chabazite, heulandite, stilbite, apophyllite and calcite. It is referred therefore to the first period of alteration.

Garnet

In several of the thin sections, numerous small grains of some mineral of high relief and hexagonal or rounded outline are found, regarding whose identification there is some slight doubt, but which may be referred to garnet with most probability. They occur normally in association with that group which is believed to have been the result of the most intense metamorphic action. In hand specimens, individual grains cannot be distinguished, but the presence of small clumps of some substance of a peculiar, light, ashy-green color in certain associations have been found to be due to a multitude of grains of this character.

One of the best examples is slide 98. The relations are sketched in Plate XIII, fig. 1. The garnet is here associated with albite, actinolite, specularite and prehnite, forming crusts on dense basalt. The grains in juxtaposition form considerable aggregates, or they are scattered in swarms through the prehnite. The average diameter of individual grains is 0.02 mm., and the maximum is not much greater. Each individual consists of a clear, colorless exterior and a darker nucleus of a brownish

color, which, under the highest powers, has a granular appearance. This opaque substance is not identifiable with certainty but is evidently some material which could not be worked over or digested in the process of recrystallization and therefore remained as an insoluble residue. It would appear that either TiO_2 or MnO_2 might act in this manner. Both are shown by analysis to be present in the basalt, and the opaque, brownish dust generally present in the garnet and at times in other secondary minerals may be referred with great probability to one or both of these oxides. The relief of the crystal grains is very high. The Becke test indicates much higher index than adjacent prehnite and therefore higher than 1.65. The characteristic outline is that of a regular hexagon. On account of the high relief, it is sometimes possible with good magnification to make out a number of faces of an individual grain and to determine that the form is that of a rhombic dodecahedron (110). In most cases, the grains appear isotropic, but occasionally a very low birefringence is perceptible. The crystalline form, high relief, habit of including foreign material, lack of cleavage and anomalous birefringence correspond with garnet. This mineral has not heretofore been recognized under exactly similar circumstances, so far as the writer is aware.

The hexagonal outline of the garnet is best preserved where the growth of the associated prehnite has made least advance. Where the prehnite reaches a more notable amount, the garnet grains become rounded, and, with good magnification, the surface appears pitted. At a further stage, the grains become smaller and finally disappear, leaving the brown dust remaining as a turbidity in the prehnite. There is some evidence that the garnet is likewise yielding to the advance of the green amphibole. The inference is that the period of deposition or field of stability for the garnet was very limited.

In slide 53 (Plate XI, fig. 5), an original glassy crust on dense basalt has been metamorphosed to a mixture of garnet, green amphibole and specularite, which has subsequently been attacked by prehnite. The insoluble residue of the original glass (TiO_2 or MnO_2) has been included as before as a muddy brown sediment in the garnet grains, giving to many of them an opaque appearance. By reflected light, this has a milky look. The spherulitic markings and curved bands, which have been spoken of before and which are illustrated in the figure, are probably surviving structures of the original glass. The garnet grains are individually rather larger than those in 98 (about 0.075 mm.) and in places are so crowded as to form considerable masses, some of which are perfectly opaque from undissolved residue. Where invasion by prehnite has progressed, the garnets are much corroded. Slide 109 is from the same hand

specimen as 53, and, so far as the relations of garnet are concerned, is almost a duplicate (Plate XIII, fig. 2).

In 115 and 118, the same relations hold. In several places in 115, the geometrical patterns in which the garnet grains are arranged show up in a remarkable manner. Plate XII, fig. 5, is a sketch of a portion of the slide. With encroachment of prehnite, the figures become obliterated. The resemblance to chloritic nodules derived from resorbed olivine, shown in fig. 2, slide 48, is striking.

In most of the slides in which garnet has been found in any notable quantity, these circular patterns are a marked feature. The resemblance in form to chlorite nodules is so close that a similar explanation of genesis appears probable. The origin of the chlorite nodules may be traced backward through intermediate steps to phenocrysts of olivine, which, it appears, became unstable during the primary consolidation of the magma and were largely resorbed. Such nuclei of olivinitic material, which had suffered refusion and partial absorption, remained unchanged in the glass after consolidation until circulating waters gained access. The time required for this varied according to the relations of the channels of circulation, and the nature of the resulting minerals depended upon this period. It appears that when alteration was delayed until a rather late stage, the nodules of resorbed olivine passed over into chlorite, but in an earlier period, while temperature was high, garnet was the chief mineral formed.

The garnet seems to alter into prehnite with great facility. Both in the derivation of garnet from olivinitic material and in its transformation into prehnite, the process appears to have followed the usual line taken in these rocks, by which magnesia and iron were reduced.

In several other slides (for example, 50, 95, 100), small grains of similar characteristics (isotropic and of high relief and hexagonal outline) are found. Occasionally they form considerable clusters, but further stages of alteration have supervened and obscured the relations. In slide 50a, Plate XI, fig. 6, clusters of them are seen to form a band in prehnite immediately adjacent and parallel to what appears at first to be unaltered basalt. In the latter, it is found, however, that the feldspars and diopside have been wholly replaced by prehnite, garnet and amphibole, while the magnetite has been left almost undisturbed and still outlines the original texture.

The Amphiboles

A number of distinctly different varieties of amphibole occur among the secondary minerals. Some of these possess decidedly abnormal char-

acteristics. Their relations among themselves and to the associated minerals present several interesting features.

Variety 1. In a hand specimen of pectolite, altered in part to crystalline calcite, small, tabular crystals of some mineral of a dark-green color were noted. A thin section (116) disclosed the relations shown in fig. 14. The dark mineral is surrounded by calcite, and between the two there is often a muddy border, indicative of decomposition. The process of replacement has frequently left the curious projecting shreds shown in the figure. The general habit and the optical properties indicate an amphibole, but several features are abnormal.

The pleochroism is strong. X = dark blue-green, Y = claret and Z = pale yellow-green. Intermediate sections show tints of gray, lilac,

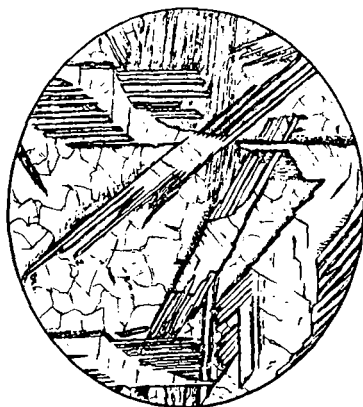


FIG. 14. Shreds of an abnormal amphibole (probably arfvedsonite) in calcite.
× 32. Slide 116.

purple and brown. Absorption formula, $X > Y > Z$. There is a well-defined cleavage, probably parallel to the prism, and probably another cleavage whose character could not be determined. Elongation tabular parallel to the prism. Optic character biaxial and negative, optic angle large. Extinction measured from the X -axis of elasticity to what is probably the vertical cleavage makes a maximum angle of 14 – 15° .

Unusual features are the reversal of the ordinary formula of absorption for amphiboles, the position of the X -axis, and the great range of pleochroism. These features agree very closely, however, with the rare variety arfvedsonite ($\text{Na}_2, \text{Ca}, \text{Fe}$), Si_4O_{12} with $(\text{Ca}, \text{Mg})_2(\text{Al}, \text{Fe})_4\text{Si}_2\text{O}_{12}$, as described by A. N. Winchell.⁴³ In the same slide, a small de-

⁴³ "Optical Mineralogy," p. 114, 1909

tached portion of the section shows calcite and quartz, with a shred of amphibole. Using the birefringence of the quartz as a basis, the amphibole gives a maximum birefringence of 0.025, which agrees very well with arfvedsonite. The possibility of ægirite was considered, but the properties agree much less perfectly with those of that mineral. Not many occurrences of arfvedsonite in this country have been noted and these only in eruptive rocks. A further study of its properties with more certain identification would be of interest. It is to be regretted that so little of it has been found. In slide 153, containing pectolite and large albite crystals, a few shreds appear to be the last remnants of arfvedsonite. In 108, a single shred shows the characteristic pleochroism (blue-green and lilac) and in 77, a half dozen remnants show pleochroism as in 116,—X blue-green, Y claret and Z yellow-green, with $X > Y > Z$. A search has been made through a large number of hand specimens in the endeavor to find other occurrences. A few cases were observed, but always so much decomposed that thin sections could not be obtained. In its typical habit, the mineral consists of groups of tabular crystals or somewhat scattered individuals resting upon or intergrown with quartz, datolite, pectolite or albite. It was always decomposed to a large degree to a grayish-green, pulverulent substance, or its total removal had left narrow gashes in the associated minerals. An interesting feature brought out was that similar gashes were not uncommon in many specimens in which there was no other evidence of arfvedsonite. Their frequent occurrence suggests that arfvedsonite may have been formed in considerable quantity at one stage of alteration but that in most cases it has totally disappeared.

Varieties 2 and 3. The usual amphibole found in the slides is of very different character. It occurs in tufts or bundles of fibres or in groups of slender prisms, and in general character resembles actinolite. It is found in small quantities in a great number of the thin sections, but in several in which the minerals associated with it are those belonging to the first periods of alteration, it is present in notable amounts. In 98, it occurs in tufts of small fibres, dispersed in columnar or plumelike forms, and at times occupying most of the field (Plate XIII, fig. 1). Pleochroism strong, from a clear emerald-green parallel to Z to pale brown tinged with green at right angles to Z. No difference in pleochroism between X and Y can be perceived. Absorption strong parallel to Z, weak parallel to X and Y. Extinction nearly parallel to elongation of the needles, but angles up to 11° were obtained. Direction of elongation makes an acute angle with the Z-axis of elasticity. Index of refraction greater than prehnite and approximating that of calcite, indicating a value of about 1.65. The maximum interference color in the section is a blue of the first order,

corresponding very closely to that of associated prehnite, and indicating a value of birefringence of about 0.033. Its affinities seem to be with actinolite. Its strong pleochrism points to a high content of ferrous iron.⁴⁴

In 53, green amphibole occurs in associations very like those in 98. Plate XI, fig. 5, shows a portion of the slide, but not that in which the amphibole is best developed. The pleochroic colors are the same, and the general appearance is very similar, but the habit is more columnar, and the plumose development is much less prominent. Extinction angles measured from the elongation gave maximum angles up to 32 or 33° (in a doubtful case still higher). Pleochroism, emerald-green parallel with Z, pale yellow and brown parallel with X and Y. Absorption corresponds. The elongation makes an acute angle with the vibration direction Z. The birefringence is practically that of prehnite, or about 0.033. Index of refraction greater than that of calcite (> 1.66). The high birefringence may indicate an approach to the iron amphibole, grünerite, but the large extinction angle and character of the pleochroism are somewhat anomalous.

The relation of the amphibole and garnet in this slide is at variance with what was found in 98 (described under garnet). In the latter, the amphibole held inclusions of the garnet and was quite plainly replacing it. In 53, it is even more evident that garnet occupies areas formerly held by the amphibole. The replacing garnet is transparent (free from opaque dust), so that the forms of the former amphibole prisms are distinctly outlined. In some instances, the replacement is not complete and remnants of amphibole are still left.

In 109, from the same hand specimen as 53, there appears to be the following relation of garnet and amphibole (Plate XIII, fig. 2). The stouter prisms of amphibole have been partly replaced by garnet. This has not proceeded very far, and a reversal of the process has occurred by which a bordering rim of fibrous amphibole has grown out among the garnet grains, partly replacing them. At the same time, the stouter prisms themselves have broken up into the fibrous form, which preserves fairly well the outward boundary but has assumed an internal structure which is even plumose in places, without much regard to the original crystallographic orientation. In the middle portion of the drawing, a single prism of amphibole illustrates both processes. At one end, the amphibole has been replaced by colorless garnet. Toward the other end, fibres of amphibole have shot out among the garnet grains for considerable distances beyond the original border of the rod. The structure of the

⁴⁴ DANA: *Op. cit.*, p. 389.

WINCHELL: *Op. cit.*, p. 109

interior of the prism has altered into masses of fibres of various orientation.

The fibrous form of amphibole with low extinction angle is found in numerous slides. In 115 and 118, it occurs in abundance. Invasion of garnet areas by amphibole is exhibited in a number of places. In 117, a series of subspherical growths of amphibole, built up of plumose or ray-like fibres, form a border between comparatively unaltered basalt and transparent prehnite. Pyrite (or chalcopyrite) is in intimate intergrowth. The pleochroism of the amphibole is much less marked in this instance. Colors vary from yellow-green, parallel to Z, to very pale bluish green or almost colorless at right angles to this. Elongation of fibres is nearly parallel to Z as before.

In the occurrences described, the very fibrous amphibole is found under circumstances which show that its formation began in early stages of alteration. Its range of stability appears to have been very extended, however. It has been observed macroscopically in small acicular tufts or sheaves resting upon the free crystal faces of heulandite, laumontite, apophyllite and calcite, minerals of a late stage of alteration, in such manner as to show later deposition. This inference is confirmed in the slides. In 72, very fresh-looking groups are found in the midst of heulandite, and in 51a, it is associated with heulandite and calcite. In 120, a group of fibres is surrounded by laumontite, and in 98 perfectly fresh amphibole is surrounded by chabazite. Slide 129 shows a sheavelike bundle of amphibole fibres in the midst of a large apophyllite crystal. In the hand specimen of the last, similar tufts were seen, resting on a free face of the apophyllite crystal. Slide 138 shows similar inclusions. In a great many other slides (50, 50a, 62, 110), small quantities are found in various associations. In some cases, there might be a question whether the amphibole might not be a survival from a previous period of more intense metamorphic activity, but when it is seen macroscopically, resting upon the faces of projecting euhedral crystals, the only inference which appears justifiable is that it is of later formation. From these relations, it appears probable that it continued stable at rather moderate temperatures. There was, however, in the more open channels, a tendency toward solution because of a diminution of iron and magnesia in the circulating waters. In the places where it is found, there appears to be always some comparatively unaltered basalt within a short distance, generally in the same slide, but in the large masses of secondary minerals sometimes found in the cavities in the trap sheet, there is no distinctly iron or magnesia mineral left. The writer has seen masses a foot or more in size consisting of pectolite, prehnite, natrolite and thaumasite lying between the bowlder-

forms of basalt, and showing apparently the complete removal of iron and magnesia compounds. The inference is that in the channels in which circulation was most free, the waters were too dilute in iron and magnesia for the deposition of these minerals, and it was only where the circulation was more obstructed and therefore slower, and where a supply of iron and magnesia was at hand in the less altered basalt, that these compounds were deposited.

The manner in which the amphibole was removed by dilution of the solutions appears in slide 108. The bundles of acicular fibres are losing their characteristic euhedral terminations and appear as small masses with irregular outline. There is no indication of alteration products, and

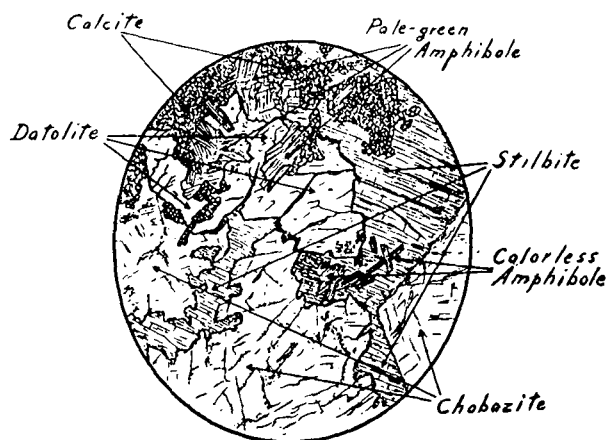


FIG. 15. Association of amphibole, datolite, chabazite, stilbite and calcite. The amphibole shows both green and colorless varieties. Datolite shows corrosion at contact with zeolites, chabazite is cut by veinlike stilbite, and granular calcite replaces all. $\times 32$. Slide 108.

it appears as if the amphibole were simply being dissolved out. Datolite and heulandite are the associated minerals.

Variety 4. Under certain circumstances, the amphibole appears to suffer a diminution in iron content without destruction of the crystals, perhaps altering in the direction of tremolite. This is observable in several slides. The green and brown pleochroic colors have practically disappeared. The crystals retain their outline and fresh appearance, but are colorless or show a very faint tinge of green. A group in slide 108 (fig. 15) shows transitions between individuals which are decidedly pleochroic and those in which the green is barely perceptible. Datolite is associated, but zeolites and calcite are the prominent minerals present.

In other parts of the slide, the amphiboles are a pale green to deep green. In several places in 94, groups of very pale green crystals are seen. In 120, 100 and 51a, similar, almost colorless crystals are observable.

Among the several varieties of amphibole present in these rocks, the arfvedsonite is believed to have been first in order of formation, the columnar prisms with large extinction angle second, the fibrous form resembling actinolite third and the colorless variety fourth.

The changes noted in the amphiboles conform to those which a complex solid solution might be expected to undergo when the external conditions are continually changing. The relations of amphiboles among themselves and to pyroxenes are undoubtedly very complex and are not well understood, but the researches which have recently been carried forward in the Geophysical Laboratory at Washington have cleared up a number of hitherto doubtful matters.

In a study of the nature of the MgSiO_3 series of minerals, E. T. Allen, F. E. Wright and J. K. Clement⁴⁵ found that MgSiO_3 exists in four modifications or phases: a monoclinic and an orthorhombic pyroxene and a monoclinic and an orthorhombic amphibole, and that the last three pass into monoclinic pyroxene at high temperatures (1150° and upward) with a slight evolution of heat. They draw the conclusion that all are monotropic toward the monoclinic pyroxene, which is the only stable phase at all temperatures. Somewhat opposed to this inference, however, is the fact, which they and other investigators record, that monoclinic amphibole is produced experimentally by heating the necessary ingredients at moderate temperatures ($375\text{--}475^\circ$) for 3 to 6 days in aqueous solutions; also the numerous examples in which the transformation of pyroxene into amphibole has been observed in nature (as in uralitization). They regard natural occurrences of amphibole as examples of the persistence of metastable phases, but this explanation seems hardly adequate to account for all the phenomena which petrographers have observed. A factor which is undoubtedly of great importance in the matter is that no member of the series ordinarily occurs pure in nature, but contains other members associated with it either in chemical combination or in solid solution. This may exert a powerful effect upon the direction of transformation under given conditions.

Further study in the same laboratory on "Diopside and its Relations to Calcium and Magnesium Metasilicates"⁴⁶ has brought out very interesting relations. The investigation was devoted to pyroxenes and did not

⁴⁵ Am. Jour. Sci., 4th ser., vol. 22, p. 385, 1906.

⁴⁶ ALLEN, WHITE, WRIGHT and LARSEN: Am. Jour. Sci., 4th ser., vol. 27, pp. 1-47, 1909.

include the amphiboles, but it may be inferred that somewhat analogous relations hold. It seems most probable that the amphiboles consist of a series of metasilicates which are capable of uniting among themselves in compounds of definite composition and in mixed crystals (solid solutions) of variable composition. In such complex solid solutions as are formed by associations of various members of the series, equilibrium with the liquid with which the crystals are in contact is necessarily easily displaced by changes in the composition of the liquid, and perhaps also by changes of temperature. The metamorphoses which the amphiboles of the Watchung rocks are found to have experienced are in harmony with this conception and would be expected from the conditions to which they were subjected.

Somewhat similar phenomena to those described are mentioned by J. P. Iddings:⁴⁷

Common hornblende and basaltic hornblende in some instances alter by the loss of color and subsequent passage into pale or colorless amphibole, resembling actinolite and tremolite; often becoming recrystallized as acicular or fibrous aggregates (strahlstein).

The chemically simpler varieties, like tremolite and actinolite, are more stable than the more complex hornblendes. In fact, the former frequently result from the breaking-down of the more complex amphiboles.

Regarding relative stability, however, the present writer would adopt a somewhat different form of expression. The more complex hornblendes are probably perfectly stable under the conditions of deposition, but with a change of external conditions they tend to change in accordance.

In the study of the transformation of labradorite into albite, it was found that the original crystals gave up to the liquid that constituent which was in excess and received that which was deficient, but with the amphiboles it appears that the process which prevailed was a breaking-up and solution of old crystals and deposition of new.

Specularite (Specular Hematite)

Specularite is seldom present in large quantity, but a small amount is often found. It has been observed without the microscope in dark blade-like crystals or groups or as a bright crimson or iridescent purplish dust on faces and as inclusions in crystals of almost every mineral in the series. The magnetite of the original rock appears to have broken up almost as soon as recrystallization began, and its place was taken by hematite. In slide 53, it is found in tabular crystals, reddish-brown by transmitted,

⁴⁷ "Rock Minerals," p. 340

blue-black by reflected light. In 98, crystals with hexagonal outlines are prominent (Plate XIII, fig. 1). One of these reflects light in such a manner that the three upper faces of a flat rhombohedron can be seen. In 61, hexagonal tablets are associated with analcite, heulandite and calcite. It is occasionally found in other slides, but no features of special interest have been observed.

During the process of ordinary weathering, hematite is not usually formed, but some hydrated compound of the sesquioxide of iron. This applies to the original basalt and to its secondary minerals. Surface decay results in the familiar rusty brown color from the iron compounds, and the hematite itself partakes of this transformation. It is quite evident, therefore, that the processes which were at work in the formation of the zeolitic and associated minerals were essentially different from weathering. The principal factor involved, so far as the hematite indicates, would seem to be difference of temperature. There is probably a fixed point at which transition occurs under ordinary conditions from hematite into hydrated forms of Fe_2O_3 , or vice versa, and the presence of hematite indicates that the temperature throughout was in excess of this minimum.

The presence of a purplish dust of hematite often adds to the beauty of crystal specimens. It occurs both as an outer coating and as inclusions, and the inclusion of hematite in a very finely divided condition may account for the delicate pink or flesh tints which the zeolites often show. There is some question, however, whether in the latter case the phenomena may not frequently be due to a ferric compound in solid solution. It has been found in the thin sections that some of the specimens which show such tints carry inclusions of ferric oxide, but on the other hand many of them exhibit nothing to which the color can be attributed. According to interpretations of mineral analyses which have prevailed until recently, the presence of ferric oxide in a zeolite would at once be attributed to a mechanical impurity. It is now recognized, however, that quite diverse compounds may enter a solid solution. A great deal of work requires to be done to ascertain the limitations of this form of combination and what interpretation is correct for individual cases.

Pyrite and Chalcopyrite

The total amount of pyrite and chalcopyrite is small, and their occurrence is of interest chiefly because of the indication it affords of the nature of the processes of alteration. Their presence is attributed to emanations from the consolidating magma and deposition in crevices. In contact with

circulating waters some migration and recrystallization may have followed, but the survival of such readily oxidizable compounds argues strongly against any oxidizing property in the waters. They have been observed with the naked eye associated with many members of the series under conditions which imply contemporaneous deposition. When surface waters have reached them, the ordinary oxidation products appear, but when seen in their original condition, they are fresh and bright.

One is apt to consider chalcopyrite as a mineral deposited from highly heated solutions, and its formation among the later minerals of the Watchung series might be considered an argument that a high degree of superheating still prevailed. This, however, is opposed to evidence from other directions and does not appear necessarily true. W. Lindgren⁴⁸ places pyrite and chalcopyrite among minerals persistent from igneous conditions up to near surface, and also believes that they were formed at times in the lower ground-water zone (zone of sulphide enrichment). Direct evidence on the formation of chalcopyrite at moderate temperatures is supplied by the observations of Daubrée⁴⁹ upon copper minerals found upon Roman coins at the hot springs of Bourbonne-les-Bains. The temperature of the water in this case was only 58–68°C. It carried in solution chlorides and sulphates of alkaline bases and of lime and magnesia, together with bromides, carbonates and silicates, and traces of various other compounds. A number of copper minerals of recent deposition were recognized: cuprite, chalcocite, tetrahedrite, covellite, tennantite and chalcopyrite. The last was recognizable both by its characteristic yellow color and by its octahedral crystalline form. Mammelonated forms were also deposited.

The minerals so far studied appear to have been formed in the first stage of recrystallization. Some of the new species were limited to a very short period of deposition, while with others the range was more extended. It is interesting to observe that all of the minerals resulting from the primary consolidation of the magma were attacked and recrystallized during this first stage of alteration. The chemical rearrangements, however, were not so complete as is implied in the disappearance of all the mineral species. Of the two molecules which made up the plagioclase feldspar of the basalt, anorthite was broken up, but albite remained stable and recrystallized as such. Several of the molecules which formed the mixed crystals of diopside probably merely suffered a change of phase in their recrystallization into amphiboles. A portion of the magnetite was

⁴⁸ Econ. Geol., vol. 2, pp. 122, 125, 1907.

⁴⁹ "Géologie Expérimentale," Paris, pp. 72-86, 1879.

split off as hematite, implying a certain amount of chemical reaction. For the formation of such compounds as are entirely new, reactions must be assumed. In this way, garnet, quartz and some of the amphibole molecules must be accounted for.

The second stage of alteration was characterized by prehnite, pectolite and datolite. Of the three, prehnite and pectolite are formed of the oxides to which the phase rule was applied in the earlier pages of this paper; that is, they belong to the soda-lime-alumina-silica series, to which quartz, the feldspars and the zeolites belong, and their relations to preceding and following members of the series are of especial interest in the inquiry as to the applicability of the phase rule. Datolite cannot be considered a member, on account of the presence of boric oxide. Its relations to other minerals are of interest, however, on account of the evidence of changing conditions which they offer.

Datolite

In the slides, the datolite is easily recognized by its moderate refraction (much higher than most of the associated minerals) and brilliant polarization tints. Prehnite is the only mineral with which it might be con-

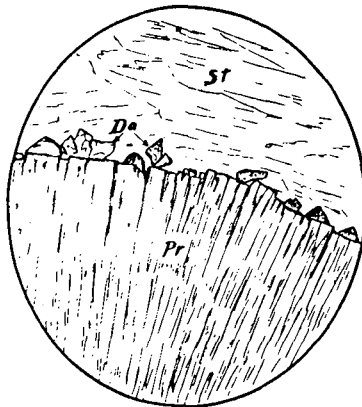


FIG. 16. Remnants of datolite crystals (Da) perched upon prehnite (Pr) in contact with stilbite (St). $\times 35$. Slide 101.

fused, but this assumes a radial development or a twinning structure which are unmistakable. In hand specimens, datolite is often seen in large, beautifully developed crystals, not infrequently an inch or an inch and a half in diameter, clear, glassy and of a light green color, showing a great number of distinct crystallographic faces. The finely granular

aggregates, however, in which it is mixed with other minerals, afford better subjects for the microscopic study of replacement phenomena.

In slide 63, in which datolite is in association with albite, as previously described, the normal appearance of the unaltered mineral is shown. It forms an aggregate of interlocking anhedral crystals, approximately equidimensional. The character is biaxial and negative, optic angle large, dispersion formula $\rho > v$. The manner in which albite is replaced by datolite in slides 63, 70 and 123 was described under albite, and the replacement of quartz in 70 under quartz. The last is shown in fig. 8.

The relations of datolite and prehnite are shown in several slides. The deposition of the two minerals is considered to have been approximately contemporaneous, on account of similar relations to preceding and follow-

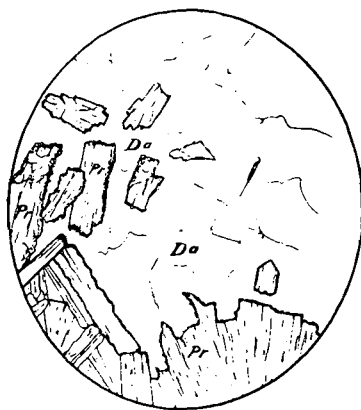


FIG. 17. Corroded crystals of prehnite (Pr) in the midst of datolite (Da).
× 35. Slide 67.

ing species, but in the cases where it has been possible to determine the relative succession of the two, datolite appears to be later than prehnite. In slide 101, small crystals of datolite are perched upon a radiate group of prehnite crystals in contact with stilbite, as shown in fig. 16. From their position, it seems necessary to suppose that they were deposited upon previously-formed prehnite. In 67, radial and columnar prehnite is mixed with datolite in an involved manner. In places, the prehnite shows strong corrosion, and outlying fragments are on the verge of disappearance. Small portions have been isolated by datolite. The apparent relations are shown in fig. 17. In 74, there is undoubted replacement of prehnite by datolite.

The relations of datolite to the zeolitic minerals seem very plain. It appears that under normal conditions datolite was in process of removal

during the entire period of formation of zeolites. Such a condition does not appear surprising, if the inferences as to the source of the boric acid are correct. The amount deposited by sublimation in the crevices of the lava would be a definite quantity, and no further supply would be available during progressive alteration of the basalt. Under the continual leaching of the uprising waters, it should therefore gradually disappear.

In section 95, the relations of datolite and chabazite are well shown. The rock is a quickly chilled basalt, in which phenocrysts of plagioclase and diopside are scattered through a groundmass of feathery microlites. It contains numerous irregular cavities which probably represent steam vents. The chief filling of the spaces is chabazite and heulandite, but along the margin of the amygdulæ are numerous corroded crystals of



FIG. 18. Datolite (Dat) replaced by chabazite (Ch) and calcite (Cal). $\times 35$.
Slide 95.

datolite. The manner in which the datolite is yielding to chabazite is sketched in fig. 18. Frequently the datolite crystals retain strong suggestions of their outward form, but the core is gone and its place is occupied by chabazite. Between the datolite and the less altered basalt, finely granular, muddy-looking calcite has entered in the manner which has been found to be so frequent.

In 110, there is a similar replacement of datolite by stilbite (fig. 19). It is noticeable in many cases, as the figures indicate, that parts of the datolite crystals are unattacked and retain their proper outlines, while other parts are deeply corroded.

In 94, there is replacement by heulandite. At one point, there is a suggestion in the structure of the datolite, heulandite and associated calcite,

and in the radial disposition of included dust, of a former spherulite in the basalt, various occurrences of which have been described.

In 75, there is probably a replacement of datolite by natrolite. Slide 108 furnishes a fine example of deeply corroded crystals in contact with



FIG. 19. Replacement of datolite (Da) by stilbite (St). $\times 35$. Slide 110.

stilbite, heulandite and chabazite. Fig. 15 shows the association at one point. With the datolite, a considerable quantity of amphibole is found. In one or two places, the basalt is very little altered and is seen to have a typically vitrophyric texture. The remnants of datolite and amphibole

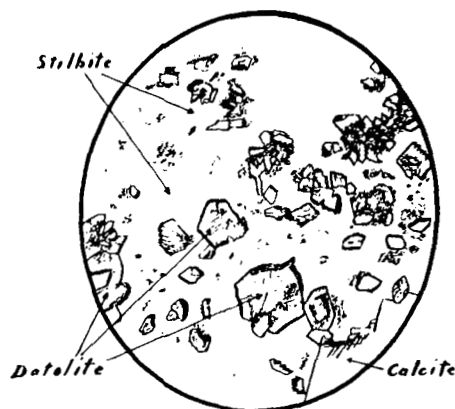


FIG. 20. Swarms of corroded crystals of datolite in stilbite. $\times 35$. Slide 83.

show a distinct tendency to form borders to the less altered rock, while the zeolites are best developed farther out. This is probably the result of leaching-out of datolite where circulation was most free.

Fig. 20 shows the appearance of swarms of corroded datolite crystals in stilbite in 83. Many of them have the rounded and etched appearance which is seen when crystals of a salt slowly dissolve in water.

Other examples might be given of the corrosion of datolite in contact with heulandite, chabazite, stilbite and calcite. In a great number of slides, there are at least a few remnants of datolite, and it may have been present originally in great quantity. The general relations and mode of replacement have been brought out in the examples given. These show that albite, quartz and prehnite were replaced by datolite, which in turn yielded to chabazite, heulandite, stilbite, natrolite and calcite. This order has been found to be true in so many cases that there is little doubt that it is general. Nevertheless, one abnormal occurrence has been observed, in which crystals of heulandite and calcite are encrusted with datolite, implying deposition at a much later period. The formation of datolite was probably conditioned chiefly by the concentration of boric acid in the solutions. In the ordinary course of events, the solutions appear to have gained access to the available supplies of this material at an early period and to have effected the reactions by which datolite was formed, while at a later period the process appears to have been one of gradual removal; but it is not difficult to conceive that under somewhat irregular conditions of circulation the boric acid might have been left until a much later period before it was taken up in the general circulation.

Prehnite

Prehnite is found in the familiar form of groups of tabular crystals, more rarely in small, distinct prismatic forms. Both varieties appear in the thin sections. The structural characteristics and optic properties are normal. The microscopic twinning lamination, which produces a plaid effect, is well shown in a number of the slides, and when seen, differentiates it immediately from all other minerals present. Equally characteristic is the deep azure-blue, low-order interference color. It is observed that when sections give the normal gray tone as the minimum interference, the dispersion may be either $\rho > v$ or $\rho < v$, but in those in which the minimum is the abnormal Berlin blue, the dispersion for red is much less than that for violet. Iddings gives $\rho > v$, in some cases $\rho < v$. Leather-brown tints are also distinctive. The interference figures rarely show distortion, except when it is evident that several individuals or groups of twinned crystals are concerned in the effect. Commonly the habit is radiating or plumose, but in two or three slides (for example, 74 and 67), it is found in small distinct prisms. The feathery forms and brilliant polarization colors often suggest bright-colored plumage.

The relations of prehnite to albite, quartz, garnet and datolite have been described. With regard to pectolite, the slides appear to present evidence that in its growth, needles of the mineral have been advanced through previously deposited prehnite, as if the latter offered no resistance. It will appear later that natrolite also, whose habit of growth is in similar slender needles, possesses the same property of advancing through previously existing crystals. In 87, finely felted pectolite lies in contact with a spherical mass of radiating prehnite. The terminations of the prehnite crystals have lost their characteristic appearance and sharpness of outline, and the borders appear muddy. In places, the felt of pectolite is plainly advancing across the prehnite.

In 76, masses of prehnite are thoroughly impregnated with pectolite needles. These cross the prehnite areas singly or in groups of three or

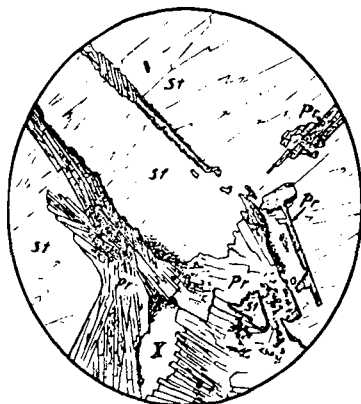


FIG. 21. Remnants of prehnite groups (Pr) in stilbite (St). $\times 35$. Slide 74.

four or lie in bundles or diverging rays. The perfectly straight and sharply defined needles pierce numerous grains of prehnite without diversion. Similar effects, though not developed so extensively, appear in 50a and are illustrated in Plate XI, fig. 6. In this illustration, the straight needles near the center represent pectolite.

In 93, the hand specimen shows grains of prehnite, the size of buckshot or smaller, which lie isolated in the midst of masses of pectolite. In the thin section, the prehnite appears turbid and between crossed nicols has a mottled look. Along the borders, it is difficult to decide what is pectolite and what is prehnite, though normally the two have a very different look. In 67, badly corroded prisms of prehnite are being replaced by chabazite.

Fig. 21 (slide 74) illustrates the manner in which originally radiate

groups have been broken up by stilbite into isolated individuals or small scattered clusters. In 101, a prehnite group is cut by veins of stilbite.

In 127 (fig. 3), which was described under albite, it is found that the prehnite clusters which had been deposited upon albite are in turn giving way to natrolite. In 68, also, prehnite is being replaced by natrolite, and portions of groups have been cut off by the later mineral.

The relations of prehnite and apophyllite are shown in 138. Small groups and fragments of prehnite are isolated by the apophyllite in numerous instances. The manner in which a vein of apophyllite cuts off a portion of a radiate group of prehnite crystals is illustrated in fig. 22.

Calcite effects striking results. Crystals appear in the midst of prehnite and develop in euhedral forms, which cut out areas of the earlier mineral as sharply as by a knife. This appears in slides 53, 62 and 76.

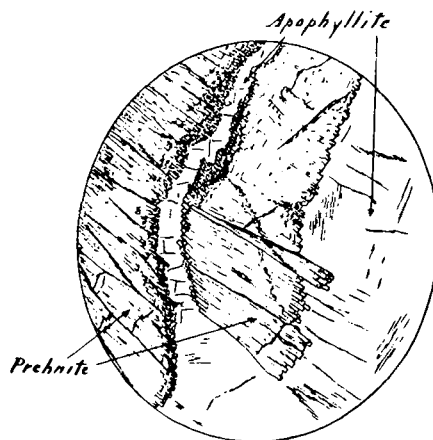


FIG. 22. Prehnite cut by vein of apophyllite. $\times 35$. Slide 138.

By a continuation of the process, fragments of prehnite of extreme irregularity are left, whose outer form is determined by calcite, but through all portions of which the same fan structure prevails.

Occasionally in specimens of basalt which appear to the naked eye perfectly normal and unaltered, small, shotlike nodules of prehnite are seen. The rock seems entirely impervious, and the relations are such as to make it appear that the prehnite had developed in the fused magma. This is so improbable that another explanation is naturally looked for, and the effects seem to be very well explained by certain phenomena which appear in several of the slides, for example, 50a, 53, 65 and 118. In 50a, crusts of prehnite border areas of basalt in which the texture appears normal at first sight, but it is found that the diopside and

plagioclase have been almost wholly replaced by prehnite, whose brilliant tints and radial structure when seen with crossed nicols give a quite unexpected appearance to the former diopside and plagioclase areas.

Although without doubt the effects of alteration and replacement are generally most pronounced immediately adjacent to cracks and crevices of various kinds, or where the solutions acted upon the glassy phase of the rock, nevertheless it appears that in places the solutions were able to penetrate by capillary action to considerable distances within unbroken rock of normally crystalline character and effect almost complete mineralogic rearrangement. At the same time, the texture was left almost undisturbed. Prehnite seems to be especially characteristic of this process, and the effects are visible in the slides mentioned. Under such conditions, large diopside phenocrysts or nuclei of resorbed olivine would partake of the transformation and would appear as prehnite nodules. A nearly circular form of radial prehnite, probably of the latter derivation, appears in slide 65.

Prehnite is determined to be later in the sequence than albite, quartz and garnet, and earlier than datolite, pectolite, chabazite, stilbite, natrolite, apophyllite and calcite.

Pectolite

Pectolite occurs in the usual groups of radiating fibres, some of which reach a length of two or three inches, but usually they are smaller. It commonly occurs in hemispherical masses. It is very frequently associated with prehnite, but it is found also with a great variety of other minerals. It is not specially well represented in the slides, but the position in the series can be fairly well determined. In descriptions of albite, quartz and prehnite which have preceded, it was found to replace these minerals. It appears under the microscope as masses of finely radiating fibres of rather high birefringence, elongated parallel to Z. It strongly resembles natrolite, but is distinguished therefrom by the higher interference colors.

In 143, the hand specimen shows an association of pectolite and natrolite in which the appearance of pectolite suggests decomposition. This inference is confirmed in the slide. The radiating needles of pectolite are buried in a mass of natrolite, the sharpness of outline has been lost and at the terminations, the crystals appear to fade out gradually, as it were, instead of terminating abruptly with sharp definition.

In 130, the hand specimen shows small clumps of pectolite fibres which are quite evidently portions of originally larger masses, buried in chabazite. The thin section was taken at the contact and shows the increasing

turbidity of the pectolite in the vicinity and the manner in which the fibres disappear in the chabazite. At one point, the pectolite area is cut across by small veins of chabazite and calcite.



FIG. 23. Masses of pectolite needles and isolated remnants of same in the midst of apophyllite. $\times 35$. Slide 129.

In the hand specimen from which 86 was cut, crystals of chabazite rest upon the hemispherical surfaces of pectolite masses. In fig. 23, the replacement of pectolite by apophyllite in 129 is sketched. Slide 135 shows practically the same relations.

General Results of the First Period of Alteration

In the slides which have been described, in which the minerals belonging to the first period of alteration are best developed, there are commonly but obscure indications as to the nature of the original material which has been replaced. Mention has been made, however, of certain circular figures and curved lines and markings, due apparently to an insoluble residue of TiO_2 , etc., upon which the forces of recrystallization had no effect. These have been interpreted as survivals of an original structure in the glass. Examples are shown in Plate XI, fig. 5, Plate XII, fig. 5, and Plate XIII, fig. 2. There are instances, too, in which the secondary minerals are separated from the basalt of normal texture by a marginal band in which the texture is of much finer grain. This is believed to represent a transition between glassy crusts and more slowly cooled interiors. In all these cases the original glass has totally disappeared, and with it such phenocrysts of diopside and plagioclase as may have been present, but in a few instances it is seen that portions of it survived.

Slide 61 is plainly a shattered glass, with whose angular fragments considerable quantities of reddish-brown clayey sediment from the underlying lake-bottom are mingled, forming a remarkable-looking mosaic. The greater portion of the glass breccia is now altered to analcite and chlorite, but in certain areas albite groups survive. Slide 119 is quite similar in origin, but alteration has proceeded to the calcite period. In many places, nevertheless, remnants of albite groups and a small quantity of pleochroic green amphibole are perceptible.

In 12, we have again a breccia mingled with sedimentary dust, but the greater portion of the glass is now represented by groups of interlocking albite crystals. The complex appearance is illustrated by Plate XII, fig. 1. In this, the lighter areas throughout are mostly albite, through which there is a sprinkling of insoluble residual dust (TiO_2 or MnO_2), distributed without regard to the crystallization of albite. The light gray in the figure is chloritic material which appears to have migrated into cracks, and the darker portions are chiefly foreign sediment. Diopside crystals are present in several places in the slide. Their outlines appear rounded, whereas the interiors have remained perfectly fresh. A little acicular amphibole is visible. The effects of later alteration are represented in a little calcite and in the bands of chlorite. In none of the slides in which the reddish sediment is present does it seem to have been affected in the slightest degree by the processes of alteration. Although it appears that the solutions reached it at the early period at which albite and amphibole were formed, nevertheless its areas and lines of distribution remain sharp and distinct.

In 120, a little albite and amphibole are scattered through zeolitic material. In one portion of the section, there are indications of an original glassy texture. In 59, the groups of minute quartz crystals shown in Plate XI, fig. 4, occur in what was originally a glass breccia. In those slides in which the geometrical figures similar to those illustrated in Plate XI, fig. 5, and Plate XII, fig. 5, appear, the distribution of minerals is suggestive of replacement of a breccia.

On the whole, the processes of recrystallization during this period are believed to have acted so vigorously upon the glass that a very small portion of that to which the solutions gained access escaped such thorough transformation that the structure was almost completely destroyed. A decided contrast is presented to the effects of the second period (that of zeolitic alteration). In the latter, it is possible to trace with ease the progress of the changes. In the less altered portions, the channels through which the solutions percolated may be followed and the differential effects upon more and less soluble material and the gradual spread of the areas noted.

Further evidence of the more intense activity of the metamorphic processes in the earlier stages is supplied by the fact that the holocrystalline phase of the basalt was undoubtedly attacked and recrystallized to a certain degree and that the phenocrysts of diopside and plagioclase left little or no trace. In the later stages, recrystallization appears to have acted only upon the glass or upon aphanitic basalts whose crystallization was of the hairlike microlitic type, and remnants of phenocrysts persist for a long time.

SECOND PERIOD OF ALTERATION

Progress of the Changes

At this point, a marked change occurs in the nature of the minerals deposited. The differences between the two periods have been ascribed, both on theoretical grounds and on the evidence of the microscopic sections, to falling temperature and to the disturbing influence of boric acid in the solutions during the first period and its elimination during the second period.

In the primary consolidation of the magma, plagioclase was present in abundance. During the first period of alteration, the anorthite molecule was immediately broken up to form new compounds to which it bore no resemblance in chemical structure. The albite molecule persisted for a short time, but soon followed a similar course. The molecular groupings characteristic of the plagioclases were destroyed. The components entered into such compounds as garnet, arfvedsonite, prehnite, pectolite and datolite. Other compounds may have been formed in the solutions, but they did not reach the point of crystallization, unless represented in minerals which subsequently disappeared completely.

During the second period, there was no further source of boric acid which could be drawn upon, and that which had been deposited as datolite was gradually taken up by the solutions and removed. The effects of the presence of boric acid disappeared. Ferromagnesian constituents also became greatly diminished. Feldspathoid compounds reappeared, and it seems as if the conditions approached those of a closed system in which feldspathoid combinations alone were present and in which the only independently variable factor of importance was temperature, which gradually diminished. As a result, the feldspathoid group of the zeolites formed the principal constituents deposited during the second period. Coincident with the change in the chemical nature of the minerals, a change in physical characteristics appears. The minerals of the first period were predominantly rather hard and of fairly high relief and

birefringence. In the second period, the characteristic minerals are soft, the refraction is less than that of balsam and the birefringence is very low.

In the glassy crusts of basalt, there is frequently a lamellated appearance, which is doubtless an original structure developed at the time of consolidation. In addition to the parallel parting-surfaces due to this feature, the glass is cut by numerous chill cracks running in every direction, so that only a little force is required to break it into a multitude of angular fragments. Definite openings of these kinds, though of capillary dimensions, doubtless offered the means for the first ingress of waters, and along these cracks, alteration first attacked the rock. This is well shown in slide 30, illustrated in Plate XII, fig. 2. Along the cracks, which show a predominant parallelism, the glass has become bleached to a much lighter color and shows a hazy birefringence. The lighter portions represent the first step in alteration and probably consist of an intermixture of zeolites and chlorite, so complete that it is entirely beyond the powers of the microscope, and no individual crystals can be perceived. It is worthy of note that among the first effects there is always a distinct bleaching, indicating removal of ferromagnesian compounds.

In addition to the openings by which the solutions were directed, there were present in the glass a number of features of physical or chemical nature which strongly influenced the course which alteration first took and traces of which remained at a stage at which recrystallization had become complete. Such were the phenocrysts of plagioclase and diopside, bubble cavities, and, perhaps more marked in their effect than all others, the nodules of olivine which were in process of resorption when solidification occurred. In the refusion of olivine, each nodule supplied a source of material somewhat different in composition from the average of the magma, and this material streamed out along flow-lines before being incorporated in the liquid. The interruption of the process by stiffening and solidification fixed and retained these differences of chemical composition. Effects of this kind may not be perceptible in the glass, but they are accentuated in the process of alteration. Brecciation of the glass and an overwhelming of the fragments in a fresh supply of lava also probably had its effect in determining differences of a physical or chemical nature and gave rise to a lack of homogeneity, which was accentuated by subsequent alteration. Various features of this kind influenced the course of alteration, and their results may be traced in many of the slides. Phenomena which are ascribed to them are shown in Plate XII, figs. 2, 3, 4 and 6, and Plate XIII, fig. 3. In some cases, the

complications are of an extreme order, and it is only by following the process through various degrees of alteration that it has been possible to arrive at satisfactory conclusions as to the genesis of the forms.

In slide 54 (Plate XII, fig. 6), the inception of the process is illustrated. The greater portion of the slide shows a fairly normal glass of a dark olive-green color, with a sprinkling of phenocrysts. Serpentinous or chloritic nodules from resorbed olivine are numerous. The attack by heated waters results characteristically in the appearance shown in the figure. The lighter bands represent lines of replacement, which often assume vermicular or crescentic forms. The course of the waters was in many cases guided by chill cracks which often show distinctly along the middle of bleached bands. Phenocrysts and spherulites have had an influence in the replacement, and there are indications that differences of composition or of physical state along flow lines, and the form of fragments of partly reabsorbed breccia were not without effect. The change consisted of a chemical rearrangement by which the soda-lime-alumina silicates passed into zeolites, and the ferromagnesian constituents were altered to chloritic aggregates or, less commonly, to serpentine.

At the point sketched, the zeolite appears perfectly isotropic and almost structureless. In other parts of the slide, similar bands show a feeble but unmistakable polarization. Analcite, chabazite and heulandite are suggested as possibilities, but specific identification is not attainable.

In 30 (Plate XII, fig. 2), the process has reached a slightly more advanced stage. In both this illustration and in Plate XII, fig. 6, a structure may be seen which is very characteristic of these incipient stages of alteration, that is, a sort of "stream effect" from the darker to the lighter areas. This may possibly be similar in origin to the stream lines seen when crystals of a soluble salt are placed in water, and is accentuated by the first stages of alteration. In other portions of the slides, the transitions from the bleached into the darker areas are by almost imperceptible gradations.

Much of the original diopside looks fresh, but it is plainly breaking up into separate granules by chloritic alteration along cracks. The small plagioclase laths retain their form, but have altered into some material giving aggregate polarization.

Slide 3 was prepared from a specimen of glass which appears to have been shattered in cooling. Along the cracks thus opened up, ferruginous sand from external sources had been deposited. Later the heated solutions followed the same lines. The resulting alteration has taken a similar course to that described for 54 and 30, but it has advanced far-

ther, and less of the original structure survives. The resulting forms appear very complex at first and would be difficult of interpretation, if clues had not been found in other sections. Plate XIII, fig. 3, illustrates a typical portion. In the upper part, one large and two small diopside crystals are seen, survivals of original phenocrysts of the vitrophyre, and at various points there are remnants of small feldspar laths. Some parts of the original glass are little altered and retain the brown or olive-green color. In other places alteration has bleached the color or has formed chloritic aggregates.

The presence of much analcite is a prominent feature of slide 3. It follows generally the vermicular courses of percolation, but in two places it lies in broad bands between lines of ferruginous sand and would seem to have filled open cracks of some width. A portion of one of these bands appears in the upper right-hand portion of the figure. The width of this band is about 0.2 mm. The other is broader, averaging 0.75 mm. The analcite includes aggregates of light-colored plumose chlorite. A small amount of non-isotropic zeolitic material is also present.

Slide 61 (Plate XII, fig. 3) is another example of a shattered glass whose angular fragments have become mingled with ferruginous sediment. The sedimentary material appears to be unaltered, whereas the brecciated glass has been changed to albite, zeolites and chlorite. Among the zeolites, analcite is prominent. There are also aggregates of hematite scales and a few grains of chalcopyrite. The forms assumed in alteration resemble those which have been described for other sections and are illustrated in the figure.

Slide 60 (from the same hand specimen as the last) shows similar phenomena. Analcite is exceptionally prominent, and other zeolites almost lacking. Chlorite and serpentine are in notable quantity. Plate XII, fig. 4, shows the characteristic structure. The banding assumed by the secondary products, which forms a prominent feature in many of the slides, is well brought out. The broad band which lies across the field a little below the center consists of a middle portion of light-green chlorite in rather large scales, bounded by darker and lighter bands of similar material in finer scales, possibly with some serpentine. Beyond, on each side, there is a sharply defined narrow band of colorless analcite, followed in most places by a band of fibrous chlorite, and more analcite in broad areas. The large circular form in the upper portion of the field is a chlorite nodule, encircled by narrow bands of analcite and chlorite. A small grain of chalcopyrite appears in the middle of the large band at the intersection with the vertical cracks.

The supposition that the bands represent original cracks is confirmed in another portion of this slide, where a very fresh-looking diopside crystal has been broken across. The two parts are now separated about 0.2 mm. and have been faulted slightly out of line, but still extinguish in unison. The faulting band is filled with chlorite and a small amount of ferruginous sediment from external sources. These cracks undoubtedly formed the passages along which the heated waters percolated, and the chemical rearrangement proceeded outward into fragments of breccia. To this process the banding must be ascribed, but it frequently reproduces the effect of a succession of layers deposited in a cavity.

Analcite

In the Watchung series, analcite was one of the first of the zeolites to appear, but under exceptional circumstances was apparently preceded by others. In the slides, it appears quite frequently, and conclusions regarding its sequence have been supplemented by macroscopic observations.

When fresh, it is perfectly isotropic and, where free from inclusions, is water-white, but with incipient decomposition it becomes very muddy. Further advance produces a porous mass, riddled with cavities. Under the latter conditions, it loses its isotropic character and becomes noticeably birefringent. Crystal outlines are sometimes lacking or very doubtful. In the fresher specimens, there appears to be a slight indication of cubic cleavage, but the fracture is in general conchoidal or follows irregular lines. Index of refraction less than balsam. Chabazite resembles analcite in its low refraction and low double refraction, but chabazite has such very low refraction that the effect of high relief is given. In addition, it has a form of blocky cleavage which is characteristic, and, in most cases, there is very little doubt as to which is present.

Analcite sometimes occurs in broad areas, as in 60 and 61, but is also apt to appear in vermicular bands. Both modes of occurrence are shown in Plate XIII, fig. 3, from slide 3. The bleached bands and cusplike figures which have been spoken of in connection with the manner in which zeolitic alteration attacks the rock are believed to consist largely of analcite, mixed probably with some lime zeolite. The transformation of albite and anorthite molecules of the glass into zeolites was probably effected with the greatest facility on account of the small amount of chemical rearrangement necessary. The first effect of such a change would probably be an intimate intermingling of species, but later, on account of the tendency which large crystals have to grow at the expense of smaller, segregation would occur, and the crystals would be individualized.

In slide 94, the chief constituents are analcite, chabazite and heulandite. The last two appear perfectly fresh, while the analcite is turbid and shows evident corrosion in contact with the other two.

In 92, the first period is represented by remnants of albite, green amphibole and datolite, which have probably replaced aphanitic basalt. The second period is represented by analcite and stilbite, with a little heulandite, and the third period by calcite. The analcite, which has become very turbid and is quite decidedly anisotropic, has been corroded and replaced by stilbite. Calcite appears to replace everything.

In 61, the analcite is found in areas of considerable size, in some places perfectly clear and isotropic and in others showing turbidity and incipient alteration. At one place, calcite is encroaching upon the analcite. In a hand specimen, large crystals of analcite are covered with a solid crust of stilbite, composed of tabular crystals disposed normally to the crystal faces of the analcite. There is no doubt of the later deposition of the stilbite.

These relations of analcite to chabazite, heulandite and stilbite are in mutual accord and appear to make the period of analcite earlier than that of either of the others. This may, however, have been true in only a limited sense. Analcite is a soda compound, whereas chabazite, heulandite and stilbite are essentially lime compounds, and the period of analcite may have overlapped those of several lime zeolites. If such were the case, a slight difference in the relative concentration of soda and lime in the solution might suffice to cause the deposition of analcite at one point, while it was being removed at another. One of the hand specimens shows a reversal of the order of analcite and heulandite, probably thus explainable. Large cockscomb crystals of heulandite have small crystals of analcite resting upon their surfaces. There seems to be no doubt that this analcite is of later deposition.

The relations of analcite and natrolite are of especial interest because of the inferences derived from the application of the phase rule. It should be found that where the two exist together, one belonged to a period of higher temperature and remained stable until a definite transition point was reached, when it began to pass over into the other. It appears that this is true and that the direction of the change is from analcite to natrolite. The relations are brought out in slide 82 (fig. 24). In the specimen from which this was prepared, small nests in a mass of porcelainlike natrolite contained crystals of analcite and laumontite, easily recognizable macroscopically. With these, natrolite crystals of much larger size than the general average of the mass were mingled. In the thin sections, the analcite appears spongy and decomposed and has

lost its normal isotropic character. The natrolite, on the contrary, looks perfectly fresh. The outline of the contact is very irregular, but it is observed that at some distance back in the natrolite, the crystals of the latter suddenly change from a finely felted to a coarsely crystalline character. The border between the two varieties is easily seen in the thin section with the naked eye and suggests the outline of an analcite crystal. It does not seem impossible that the natrolite replaced various minerals (of which laumontite and analcite were two), and that the coarseness of crystallization depended upon the facility with which replacement was accomplished. In portions of the slide where it is plain that laumontite is being replaced, the crystals of natrolite are of a fine character, but where there are reasons for supposing that analcite has been replaced,

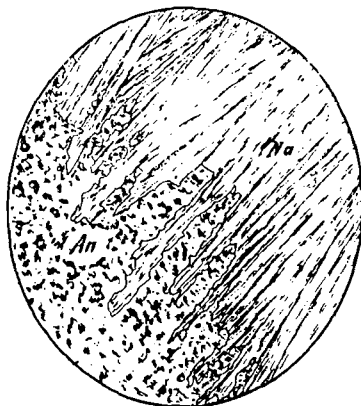


FIG. 24. Porous, decomposed analcite (An), penetrated by needles of natrolite (Na). $\times 35$. Slide 82.

the natrolite crystals reach dimensions many times those shown in the first variety.

Slide 124 is from the same hand specimen as 82 and shows essentially the same relations. In 73b, there is again a sudden increase in the coarseness of crystallization of natrolite along definite boundaries. Within the masses of larger crystals, there are two extremely irregular patches of a nearly isotropic, spongy mineral which appears to be analcite. This is confirmed in another portion of the slide, where similar forms still show crystal outlines. Replacement of analcite by calcite is also shown. In 68 also, a small group of isotropic crystals in natrolite show in places typical outlines of analcite. Portions are corroded, and replacement by natrolite is evident.

In a hand specimen, there is an association of analcite and natrolite in what appears to be a reversal of the usual conditions, that is, analcite crusts appear to rest upon natrolite. It is found, however, on close examination, that the analcite crystals are porous and drusy, while the natrolite is fresh, and the crystals show characteristic pyramidal terminations. They grow up among and probably through the analcite. The specimen is a good example of the deceptive appearance which replacement relations often assume.

The observations of Brögger⁵⁰ upon analcite found as a mineral of late development in the Norwegian syenite-pegmatite veins are of interest. He finds that analcite is the oldest and most widespread of all the zeolites found in open vugs and continues his description as follows:

As Lemberg has ascertained, analcite is formed very easily at the expense of various soda-rich silicates at somewhat high temperature. Friedel and Sarasin⁵¹ obtained beautiful crystals of analcite by heating the ingredients of albite with water up to 400°; A. de Schulten⁵² obtained, by 18 hours' heating of a solution of soda silicate or of soda in closed tubes of an aluminous glass at 180°-190°, analcite crystals up to 0.1 mm. in size, also⁵³ by heating (during 17 hours up to 180°) soda silicate and soda aluminate in the proportions corresponding to the composition of analcite in lime-water in closed copper tubes. We recall further that among the soda-rich zeolites of Plombières observed by Daubrée (where the thermal waters, which have here deposited zeolites in recent times, show only about 70°), analcite was not found, so we may almost venture to estimate the temperature at which the abundant deposition of analcite in our veins ensued at more than 70°, perhaps at about 200°.

The high temperature which Brögger assumes for analcite deposition hardly appears justified for the Watchung rocks, but a considerable degree of superheating may well have existed.

Chabazite, Heulandite and Stilbite

Chabazite, heulandite and stilbite are so frequently associated, both in hand specimens and in the slides, that they may very well be considered together. In the larger specimens, they are among the most abundant minerals and frequently form showy groups. The chabazite crystals are usually of a light salmon or salmon-pink color, and in general show only rhombohedral faces (or pseudorhomboidal, if this view be taken). Heulandite sometimes exhibits pink tints but more frequently is almost colorless. Single crystals occur, but grouped forms with curved faces,

⁵⁰ *Zeitschr. für Kryst. und Min.* vol. 14, p. 169, 1890.

⁵¹ *Compt. rend.*, vol. 97, p. 290, 1883.

⁵² *Bull. de la soc. min. de France*, vol. 3, p. 150, 1880.

⁵³ *Ibid.*, vol. 5, p. 7, 1882.

frequently in cockscomb growths, predominate. Stilbite is prevailingly of a light straw color, and the usual sheavelike clustering is the common mode of growth.

In the thin sections, heulandite and stilbite bear such a close resemblance to each other that great care must be used to distinguish them. There are two tests, however, depending upon optical characteristics, which are of such a nature that one is especially applicable to those sections in which the other fails to give good results. The first is that heulandite is positive in character and stilbite is negative, but in sections in which the lamellar cleavage is most apparent, this test fails. In such sections, however, the direction of the plane of the optic axes can usually be ascertained. In heulandite, this plane is perpendicular to the cleavage, but in stilbite, it is parallel.

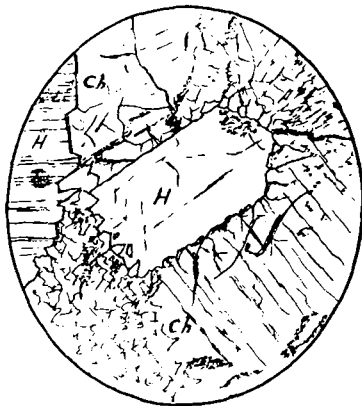


FIG. 25. Replacement of chabazite (Ch) by heulandite (H). $\times 32$. Slide 95.

There is some variability in the size of the optic angle in both heulandite and stilbite. It is never large but may become very small. In 121, a section of stilbite gives practically a uniaxial cross. The positive or negative character, however, does not seem to vary. In chabazite, the figure is usually biaxial and always positive so far as tested. In some crystals, the interference figure is distinct and the optic angle rather large, but, in many, the birefringence is so weak that it is difficult to perceive any figure whatever.

The relations of chabazite to heulandite are shown in a number of slides (for example, 95, 108, 94, 51a, 77 and 128), and it appears that chabazite always precedes heulandite. With growth and advance of the later mineral, the chabazite breaks down along the border in a characteristic manner. An example is shown in fig. 25 (slide 95). In the upper

left-hand portion of the sketch, the chabazite retains its normal outline, but at most places along the border it has a brecciated appearance. This feature is observable in a great number of instances.

The replacement of chabazite by stilbite usually takes a similar form. It is well shown in slides 110, 108, 80 and 121. In slide 108 (fig. 15), the chabazite is cut by distinct veins of stilbite, portions of which are shown in the sketch. The irregular boundaries of the veins are probably determined both by differential solution of chabazite and by the crystallizing tendencies of stilbite. In fig. 26 (slide 121), also, stilbite is advancing into chabazite in irregular veinlike form.

In slide 110, isolated crystals of chabazite surrounded by stilbite show deep corrosion. At one point in the slide, the phantom forms shown in

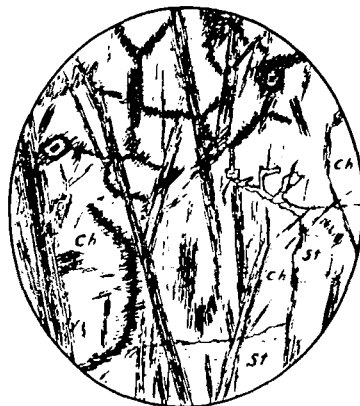


FIG. 26. Replacement of chabazite (Ch) by stilbite (St) and of both by natrolite. The natrolite needles have shot forth with apparently little opposition across areas of chabazite and stilbite, and they have also worked in along cleavage lines of chabazite. $\times 35$. Slide 121.

fig. 27 appear. These are simply inclusions in stilbite, but the forms outlined are plainly those of preëxistent crystals, which are believed to have been chabazite.

Heulandite and stilbite frequently occur together, but it is rather rare for either to show unmistakable corrosion at the contact. In slide 108, however, the evidence is fairly good that heulandite inclosed within stilbite is being replaced by the latter. In 110 and 92 also, some confirmatory evidence is found. The more usual form of contact is shown in 88, where it is perfectly sharp. In several hand specimens, more indubitable evidence is given. The heulandite appears fresh and glassy but is crusted with stilbite. The inference from the phase rule is that heulandite and

stilbite cannot coexist in equilibrium except at one fixed temperature, which represents a transition point. Although heulandite might very well persist in an unaltered form beyond this point, it should never be deposited from solution simultaneously with stilbite.

The replacement of chabazite, heulandite and stilbite by natrolite seems to have been effected with great ease, natrolite needles shooting out across areas of the three lime zeolites as if no obstacle were offered. The manner of growth as regards chabazite and stilbite appears in 121 and is illustrated in figs. 26 and 28. In the hand specimen, a polished face was prepared, and it could be seen that the chabazite and stilbite were filled with slender filaments of natrolite radiating from a large area at one side.

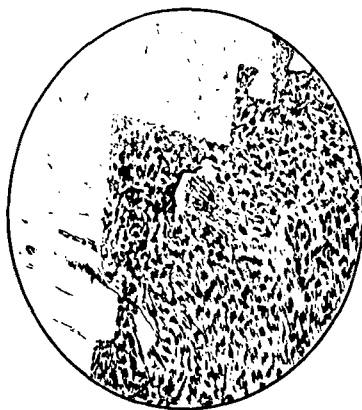


FIG. 27 Phantom crystals of some replaced mineral (probably chabazite) found as inclusions in stilbite, which extends uninterruptedly across the areas. $\times 35$. Slide 110.

In the microscopic section, the effects are similar. The slender needles of natrolite cross areas of the older minerals and also work in along joints of chabazite.

A similar replacement of stilbite by natrolite appears in 71 and of heulandite by natrolite in 70. These two are from the same rock. In both cases, crystals of the earlier minerals have become isolated in masses of natrolite. The characteristic outlines have been lost, and the borders are so impregnated with natrolite fibres that one mineral seems to fade into the other.

In 84, the hand specimen consists largely of dense white natrolite, in which small pinkish areas have a geometrical appearance and suggest that some mineral has been replaced. In the thin section, several of these areas appear. It is seen that the pink color is due to separated grains of

hematite, while the areas thus outlined have the form of chabazite, although wholly occupied by interlocking natrolite crystals. In 94 also, natrolite is seen to be advancing across chabazite.

Several hand specimens show a distinctive method of replacement of chabazite by laumontite. The laumontite appears to start from some nucleus within a solid crystal of chabazite and grow in radial crystals, until there may be only a crust of chabazite left. Slide 147 contains several radial groups of this kind. In 77 also, there is replacement of chabazite by laumontite, though in neither of the thin sections does the evidence appear as decisive as in large specimens.



FIG. 28. Needles of natrolite which have penetrated areas of stilbite. $\times 35$.
Slide 121.

In 120 and 128, there is an association of laumontite and heulandite. The relations are not as clear as might be desired, but laumontite appears later.

Replacement of all three minerals by calcite is seen in numerous instances. It appears that chabazite, heulandite and stilbite followed each other in this order and that they were all earlier than natrolite, calcite and laumontite.

Laumontite

Because of the friable nature of laumontite it was found difficult to get good microscopic sections showing it. There are only a few, therefore, which illustrate its occurrence, and in these the relations rarely appear of a decisive nature. On the other hand, fortunately, it is one of the few minerals which occur in the large specimens in such a manner as to make the sequence beyond question. It is frequently seen resting upon drusy

chabazite crystals, and the manner in which it replaces chabazite has been described.

In slide 120, it appears to be replacing heulandite, and in a great many hand specimens, it is seen to have been deposited upon previously formed heulandite. In one or two instances, it has been found resting upon stilbite.

In 82, it is found in contact with natrolite, and it is quite certain that natrolite crystals are advancing through the laumontite. On the other hand, in a large specimen, the appearance is decidedly in favor of the interpretation that laumontite is later than natrolite. It is probable that the relations of laumontite and natrolite are similar to what was found for heulandite and analcite, that is, the periods of the two overlap, and relations in a given case depend upon slight differences in concentration of lime and soda.

Similarly, in 128, it appears that calcite replaces laumontite, but in a great number of hand specimens, laumontite groups are perched upon calcite crystals. From this last occurrence it appears that laumontite forms one of the end members of the zeolite series. The period did not come to a sudden end, but a transition period intervened, during which both laumontite and calcite were deposited.

Scolecite

In a number of slides (89, 73b, 68, 71, 143), remnants of some mineral are found which is believed to be scolecite. It gives inclined extinction and is probably triclinic. Birefringence about 0.007, refraction > natrolite and < balsam, elongation negative, optical character indeterminate.

It has been found only in association with natrolite, which replaces it in the manner shown in fig. 29. Its exact position in the series is therefore uncertain.

Natrolite

Natrolite is one of the most abundant zeolites and is found in a great many of the thin sections. There is little to add to what has already been written, for the reason that, like laumontite, it is one of the end members and replaces practically everything else, and these replacements have been described under the corresponding minerals. In its relations with calcite it appears to differ from laumontite, for wherever the two come together, the natrolite always appears earlier. The replacement is well illustrated in 121. Natrolite needles which have advanced through stilbite and chabazite are wiped out abruptly by encroaching calcite.

In hand specimens of natrolite, several features are notable. It is often observed that a finely felted variety changes abruptly to one more coarsely crystalline. The border between the two frequently has a geometrical pattern. There may also be a deposition of hematite dust within the figure outlined, which produces areas of a pinkish color in the midst of

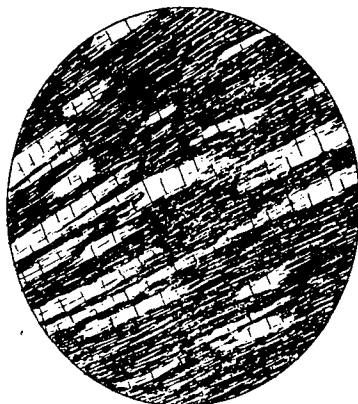


FIG. 29. Remnants of scolecite needles in the midst of natrolite. $\times 35$.
Slide 143.

solid natrolite. At times, small cavities of similar appearance are lined with projecting natrolite needles. These features are interpreted as attributable to the solution and removal of some preëxistent mineral. Generally, this has gone on *pari passu* with the growth of the natrolite, but in some cases it has been more rapid, leaving cavities for the development of well-formed terminated crystals.

Apophyllite

The replacement of various minerals by apophyllite in several of the slides has been described (figs. 9, 22, 23). From these relations, but more especially from its relation to calcite, its period of deposition is believed to be among the late members of the zeolite group. In slide 138, it is found in contact with calcite, which appears to be encroaching upon it, but on the other hand, it has been observed in hand specimens apparently resting upon calcite. It is probably one of those minerals whose period overlaps that of calcite.

Ordinarily, a small amount of fluorine is found in apophyllite (on the average about 1.5 per cent). It is possible that in our rocks, this can be ascribed to some preëxistent mineral, but it seems more probable that it was derived from a small quantity locked up by the magma when it

stiffened into a glass and was subsequently set free by the decomposition of the latter by solutions.

Apophyllite is the first mineral met in the series in which potash plays a prominent part in the formula. The source of the potash, however, is not far to seek. The analyses of typical examples of basalt given on an earlier page show potash to be universally present, and while the earlier-formed minerals are essentially soda and lime compounds, some of them undoubtedly carried a certain amount of potash replacing a portion of the soda.

Chlorite

Chlorite is almost universally present. In those slides, however, in which the minerals are prevailingly of the first period of alteration, the quantity of chlorite is so small as to lead to the suspicion that it does not properly belong here. Throughout the second period, however, it seems to have been developed in considerable quantity and to have extended through the third period likewise.

Considerable variation in appearance and properties is observable, and a number of members of the chlorite group may be present, but it has not been considered a favorable opportunity to make a detailed study to determine this. The color varies from bright emerald-green to pale yellowish green, or almost colorless. Birefringence is never strong, but varies from moderate to almost isotropic. The general habit is aggregates of minute scales, but it may develop in fairly large blades, especially when in nodules from alteration of resorbed olivine. There is quite a tendency to become segregated in definite areas or to migrate into cracks.

The constant tendency for magnesium and iron to be leached out throughout the whole process of alteration has been mentioned in several places. It was observed that the green amphibole was usually found in close association with the less altered basalt, while in the less obstructed channels it was seldom found. This relation is probably even more characteristic of chlorite. One exception has been noted, however. It is not uncommon to find vugs lined with calcite crystals which show small spheroidal groups of chlorite perched upon them.

Green amphibole is often found in the same slides as chlorite but seldom in intimate association. There is little to indicate an alteration of one into the other.

Serpentine

The quantity of serpentine is so small as to be almost negligible. It is probably present in some cases, associated with chlorite.

THIRD PERIOD OF ALTERATION

Calcite

Calcite occurs in quantities almost equal to all the other minerals combined. In habit, it is very variable. The most common form of crystal is probably the unit rhombohedron, but slender scalenohedrons (dog-tooth spar) are frequently found, and complex combinations of crystal faces are not unusual. It is also frequently developed in finely granular masses. Its presence is so nearly universal that it is rather rare to find a specimen of any mineral or group of minerals which does not show the presence of calcite, if tested with acid. In the earlier part of the work, it was found a hindrance, because of the frequency with which it obscured the study of desired relations. It was finally found necessary to apply the acid test in every case before making sections, in order to make sure that the later introduction of calcite had not destroyed the desired evidence. Its frequent presence necessitated the rejection of a great quantity of otherwise available material, and in some cases required that a long search be made before specimens free from calcite could be found.

Reference has already been made to the manner in which granular calcite of later growth frequently develops between crusts of earlier minerals and the less altered basalt and simulates earlier deposition. At times, this takes the form of well-developed crystals, upon which the older crusts appear to rest, but in such cases, microscopic examination frequently shows certain abnormalities in the relations.

In the great majority of cases, there can be little doubt that calcite is of later formation than all the minerals of the feldspathoid series, with the exception of laumontite. Occasionally its deposition may have begun somewhat earlier, but this was a rather abnormal condition, and in general, calcite is the replacing mineral.

It was found difficult at first to account in a satisfactory manner for the introduction of such quantities of carbonic acid as the abundance of calcite requires. The writer was disposed to look for its sources in vapors evolved from the magma, but although such vapors were probably given forth, they would, like aqueous vapors, have opportunity under the conditions of consolidation to dissipate freely in the air, unless they were locked up in some nonvolatile combination until the late period at which calcite appeared. Search was made for evidence of some intermediate mineral to whose decomposition the carbonic acid of the calcite could be ascribed, or for indications of an early appearance of calcite, but evidence in both directions appeared to be entirely lacking.

The possibility was also considered that while the majority of the minerals originated through the form of circulation described, the formation of calcite should be assigned to a far later period, in fact to the present cycle of events, in which cold surface waters carrying CO_2 in solution leach directly downward. The vital objection to this conception is that such waters would also be oxidizing in nature, and the associations of the greater part of the calcite render this idea very improbable.

The hypothesis which was finally adopted as being most probable was that the meteoric waters of the general form of circulation, which has been accepted as responsible for the series of earlier minerals which we have considered, retained some store of the originally dissolved carbonic acid (probably as bicarbonate of lime, $\text{CaH}_2(\text{CO}_3)_2$, and carbonates of the alkalies) at the period of their history at which they entered the basalt sheet on the return journey to the surface. When conditions became favorable, this supply of dissolved carbonic acid reacted with the various original and secondary lime compounds and gave rise to the formation of calcite. Carbonation, as is well known, is one of the most common effects of the action of meteoric waters upon lime silicates at moderate temperatures. When the temperature is high, on the contrary, the process is reversed, so that carbonate rocks carrying silica, passing from the zone of katamorphism into the zone of anamorphism, where high temperatures prevail, have the carbonic acid driven off, while the lime combines with silica and produces lime silicates. The process is, therefore, evidently an easily reversible one under variations of temperature. This probably accounts for the observed fact that little or no calcite appears to have been formed in the Watchung series during the prevalence of the more elevated temperatures under which the minerals of the first and second periods were deposited, and it was not until very moderate temperatures were reached that carbonation became the chief feature, tending to destroy the previously formed silicates. Naturally, no sharp border-line can be drawn between the periods. For a time more or less prolonged after the deposition of calcite had begun, the formation of silicates continued. Laumontite and apophyllite were deposited simultaneously with calcite almost beyond question. It is not impossible that some slight deposition of calcite began at an earlier stage, but decisive evidence of this is lacking. The fact must not be overlooked that the temperature at which deposition of calcite began would be a function of the concentration of the carbonic acid in the solutions. Under the conditions which seem to have existed in our rocks, the quantity of carbonic acid was probably minute, and the large amount of calcite found must be referred to the continued passage of the water for a prolonged period.

Under different conditions, a much larger quantity of CO_2 might be present, and the formation of calcite would begin at an earlier date. For example, in the not unusual zeolitic deposits in which the minerals form distinct veins cutting sheets of basic eruptives, it is extremely probable that circulation is generally begun while emanations are still being given forth by the cooling magma and that these are added to the ascending waters. Under such circumstances, the concentration of CO_2 might be supposed to reach a much higher value, and as a consequence, the period of calcite deposition would be much advanced. In the zeolitic veins cutting the great intrusive sill of Palisade diabase, it is observed that large crystals of calcite are encrusted with analcite and stilbite. An earlier period for calcite is indicated, which may thus be accounted for. In specimens from other localities, also, with whose geological relations the writer is not familiar, an early period of deposition of calcite has been observed.

Thaumasite and Gypsum

At about the same period as calcite, a small amount of gypsum was deposited and also the unusual mineral thaumasite. The latter is apparently a combination of silicate, carbonate and sulphate of lime. Its occurrence has been noted in only a few localities throughout the world. Gypsum has been observed resting apparently upon calcite crystals, and masses of thaumasite are found to contain nodules of pectolite, heulandite and apophyllite, imbedded like raisins in a pudding. These relations indicate a late period of deposition for the two minerals. The sulphates necessary for their formation can likewise be considered referable to meteoric sources.

During the calcite period, green amphibole, chlorite, specularite and probably pyrite and chalcopyrite were also deposited as has been described.

The presence of gypsum affords an approximate mark on the thermometrical scale. Van't Hoff and his associates⁵⁴ have made elaborate investigations on the relations of gypsum and anhydrite. They fixed the transition point between gypsum and natural anhydrite at 63.5°C. and 175 mm. vapor pressure. The presence of other salts in solution somewhat affects the value of the transition point. In a saturated solution of NaCl , they decided upon a mean value of 30° for the temperature of formation of anhydrite from gypsum. In our solutions, not all the conditions are known, but it seems certain that if the temperature at this time had exceeded 63.5°C. , the sulphate of lime would have been deposited in the form of anhydrite instead of gypsum.

⁵⁴ *Zeitschr. phys. Chem.*, vol. 40, pp. 257-306, 1903.

III. COMPARISON WITH OTHER DEPOSITS AND GENERAL CONCLUSIONS

With these minerals, the processes of alteration through ascending waters appear to have been brought to a close. This may perhaps be considered due to any one or to a combination of several causes. The waters may have reached a temperature which was practically the average climatic temperature of the region and were therefore unable to effect further changes, or the circulation may have become much enfeebled because of diminution of the stores of heat-energy in the rock, or the velocity of reaction at these lower temperatures may have become so small that its effects are negligible. The writer is inclined to believe that all of these causes cooperated.

The general order in which the minerals appeared is shown in the accompanying chart (page 179). The question arises as to the extent to which this sequence should be considered an invariable one applying to all zeolitic deposits. In considering this, the first point which must be kept in view is that the conditions under which the formation of minerals occurred in other deposits must have been similar in order that the results should be comparable. A frequent occurrence of zeolites is in fissure-veins cutting masses of intruded igneous rock, as in the Palisades. In such cases, sublimates given off by the magma at the same time that processes of alteration are in progress (such as boric oxide, carbonic acid, fluorine vapors, sulphur compounds and possibly others) and passing into the channels of circulation must affect most decidedly the periods of deposition of the compounds into which they themselves enter. Such minerals as datolite, calcite, apophyllite and the metallic sulphides might therefore appear at periods somewhat different from what has been observed in the Watchung minerals.

It has been shown, too, that the presence of boric acid in considerable quantity acts as a disturbing factor, which affects the nature of the other minerals deposited, and such other emanations as are capable of uniting with soda and lime would probably have similar effects. On the whole, however, it appears that the zeolites proper would usually follow a similar sequence in such fissure deposits to that observed in the Watchung rocks.

In zeolitic formations in surface flows of basalt, which have been brought about by a similar uprising of meteoric waters, the processes should normally be very similar, but it appears that in some instances interruptions occur. From the studies of Whitman Cross and W. F. Hillebrand⁶⁸ on the minerals from the basalt of Table Mountain at Golden, Colo., it appears that the zeolitic deposits there occur both in fissures and in

⁶⁸ Bull. 20, U. S. Geol. Surv. 1885.

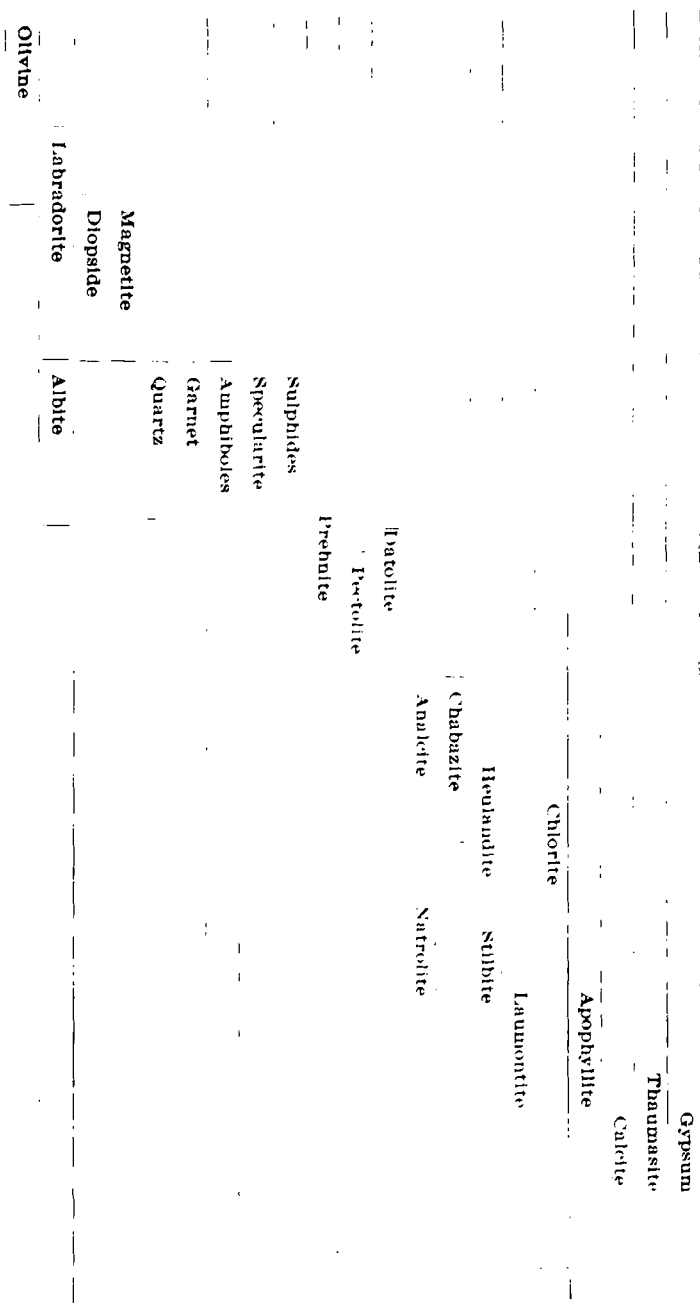


Chart Showing Order of Dependence and Periods of Stability of the various Primary and Secondary Minerals

roundish cavities in an amygdaloidal zone near the surface of the older of two basaltic flows. They may be divided into two groups, according to mode and time of formation. On considering all the species which occur in relations which permit inferences to be drawn as to relative age, the following sequence was determined:

- | | |
|----------------------|--------------------------|
| 1. Laumontite. | 7. Thomsonite. |
| 2. Stilbite. | 8. Analcite. |
| 3. Thomsonite. | 9. Apophyllite. |
| 4. Calcite (yellow). | 10. Calcite (colorless). |
| 5. Stilbite. | 11. Mesolite. |
| 6. Chabazite. | |

It will be observed that this order is different from that which has been found in the Watchung sheet, but it is seen at once that several species are repeated at intervals, implying irregularities in conditions which would have to be explained before comparisons could be made with other localities in which the sequence is normal. In such cases as this, we have, perhaps, a reversal of conditions by which the circulating waters which should, in the usual course of events, gradually become cooler, are again highly heated.

A somewhat similar condition is implied in the zeolitic deposits occurring in the copper-bearing amygdaloids of Lake Superior. The sequence here as determined by Pumpelly⁵⁶ was as follows:

The chlorite of the melaphyre, and consequently the distinctive character of that rock, is due to the alteration of hornblende or pyroxene. This seems to have been the first step toward the production of melaphyre proper. Laumontite . . . appears to have been formed either contemporaneously with the chlorite, or as the next step in the process.

The next step appears to have been the individualization, in amygdaloidal cavities, of nonalkaline silicates, viz.: laumontite, prehnite, epidote, respectively, according as the conditions favored the formation of one or the other of these.

Following these came the individualization of quartz in these cavities.

Perhaps we may be warranted in considering these minerals, together with the lime of the calcite that more rarely occurs in this portion of the series, as chiefly due to the decomposition of the pyroxenic ingredient of the rock.

So far as we may infer from the tabulated results, the concentration of copper in the amygdaloidal cavities does not appear to have begun till after the formation of the quartz.

In this part of the series falls also the formation of a chloritic or green-earth mineral, which in some manner has displaced prehnite, quartz, calcite, and with which copper, when present, appears to stand in intimate relation.

⁵⁶ "Paragenesis and Derivation of Copper and Its Associates on Lake Superior," *Am. Jour. Sci.*, 3rd ser., vol. 2, pp. 188-198, 243-258, 347-355, 1871.

Subsequently to this came the individualization of the alkaline silicates, viz: analcite, apophyllite, orthoclase. Here also seems to belong the formation of datolite. . . .

The fact that calcite occurs at almost every step in the paragenetic series and forms one of the most common of the secondary minerals is proof that carbonic acid was very generally present throughout the whole period of metamorphism; it was probably the chief mediating agent in the processes, without being sufficiently abundant to prevent the formation of silicates.

In this series again, the sequence is quite different from that observed in the Watchung rocks. Pumpelly, however, has hardly touched upon the question of the conditions under which the secondary minerals originated, especially as regards attitude of the beds, and those who have paid some attention to these matters have considered it rather from a theoretical standpoint than from internal evidence. Portions of the copper-bearing series are now buried to a depth of several thousand feet, and an unknown thickness has been removed by erosion from their upper surface. Such facts as the not unusual paramorphic change of laumontite into orthoclase might be considered to imply a reversal of the progress of alteration brought about by deep burial subsequent to a cycle of alteration in a more superficial zone. A detailed investigation of these deposits, which would include a review of former evidence in the light of more recent knowledge and a consideration of additional evidence brought to view in the depths to which mining operations have now penetrated, would be most interesting and valuable in throwing light on the difficult question of the origin and circumstances of precipitation of the native copper. Until more information is available regarding the conditions of deposition of the secondary minerals in the Lake Superior rocks, they will hardly afford a basis of comparison as regards sequence with the Watchung series.

There are undoubtedly numerous zeolitic deposits in which conditions of formation were closely analogous to those prevailing in the Watchung rocks, but, as was remarked in the early pages of the present paper, few observers appear to have made detailed observations regarding the sequence in which the minerals were deposited. Instances have been met also, in going through the literature, which bear internal evidence of rather hasty judgment in drawing conclusions as to sequence. This does not appear surprising from the misleading character of the evidence encountered in some instances in the study of the Watchung rocks, which has brought into prominence the necessity of constant caution.

In Hintze's "Handbuch der Mineralogie" a compilation is made of a great number of occurrences of the zeolites, with short summaries of the

relations which they bear to each other and to other minerals. In such cases as observations on sequence have been made, the order appears to correspond in most instances to that observed in the Watchung series. There are a number of instances recorded, however, of pseudomorphs of minerals which, from observations of the Watchung series, should be of early formation, but which have taken the form of zeolitic species. Prehnite and orthoclase appear especially likely to do this. This may possibly be due, as was suggested before, to a reversal of the normal sequence by a change of conditions due to external agencies. On the other hand, the data available are not sufficient to exclude the possibility that this was a normal order under the conditions of the case and that such factors as the composition of the original rock may enter in determining the sequence of deposition.

In discussing in previous pages the application of the phase rule to the problem and again in describing analcite, the inference was drawn that analcite and natrolite should possess a transition point, at which single point only they could coexist in equilibrium; and the observed relations of the two were found to confirm this conclusion. Two instances taken from Hintze⁵⁷ add further evidence: On the Kunetitzer Mountain, Bohemia, analcite crystals are found

up to 12 mm. in size in cavities of the basalt; often porous and covered with natrolite;

and again in the Tyrol

at the Clpit Bache at the north foot of the Schlern-Gebirg, milk-white to flesh-red crystals, scarcely more than 1 cm. in size, covered with apophyllite, more seldom with calcite and natrolite-needles; many crystals porous and altered to an aggregate of small needles (? natrolite).

Heulandite and stilbite are believed to possess a similar transition point. The well-known German mineralogist, Breithaupt, devoted special attention to paragenesis of minerals. His work dates back sixty years or more but as far as macroscopic observations go appears to be accurate and reliable. In regard to heulandite and stilbite, he records the sequence (a) heulandite, (b) stilbite, and remarks:⁵⁸ "This paragenesis is followed with perfect constancy in many localities of occurrence." It should be noted that in Breithaupt's usage, stilbite was called "desmin" and heulandite "stilbit." He gives numerous examples of the order observed for various other zeolites and for quartz, prehnite, datolite and calcite

⁵⁷ *Op. cit.*, p. 1717.

⁵⁸ "Die Paragenesis der Mineralien," Freiberg, o. 105, 1849.

from many localities.⁵⁹ In nearly all cases, they agree with that which the author has determined for the Watchung series.

Breithaupt arrived at certain well-founded conclusions regarding the conditions of formation of zeolites,⁶⁰ as follows:

It is notable that it (that is, their formation) occurs not only in the younger eruptive rocks, in whose bubble cavities the zeolites are especially at home, . . . but also in much older varieties of rock. But where the zeolites also appear, they can be observed as always only products of leaching, and have depended upon lateral secretion, and herein they accord perfectly with the occurrences in bubble-cavities. The bubble-cavities, as well as the veins in which they appear, are mostly wholly free from magnesia or iron-oxide containing minerals; at the most, only traces of these ingredients occur, whose nonexistence is related to the mineralogical-chemical character of the zeolites.

In a recent article appearing in the *Annals of the New York Academy of Sciences*, vol. xix, pp. 121-134, 1909, W. G. Levison has recorded his observations on the sequences of the minerals of the Newark igneous rocks at various New Jersey localities. His results indicate that he found no fixed order in the sequence in which the minerals appeared even in specimens from the same locality. His work, however, was done without the aid of the microscope. The present writer's investigations have indicated that replacement phenomena in these rocks have followed such a course that it is hardly possible to reach satisfactory conclusions from macroscopic observations alone.

As a final citation, which bears upon both the sequence of deposition and the general nature of the process, Brögger's well-known paper on "Die Mineralien der Syenitpegmatitgänge der Südnorwegischen Augit-Nephelinsyenite"⁶¹ is so interesting and instructive that the writer desires to quote certain parts at considerable length.

Brögger takes up the geological relations in the following order:

1. Phase of magmatic consolidation.
2. Chief phase of the pneumatolytic minerals. These are rich in fluorine, boron and sulphur, whose derivation he ascribes to the magma.
3. Phase of zeolite formation.

Regarding this phase, he says in part.

Under the continued cooling of the veins and their surroundings, the conditions of mineral formation must by degrees change more and more: finally the pneumatolytic exhalations, which at first must have consisted principally of fluorine, chlorine and boron-rich combinations, then of sulphurbearing combi-

⁵⁹ *Op. cit.*, pp. 103-107, 260-261

⁶⁰ *Op. cit.* p. 259

⁶¹ *Zeitschr. f. Kryst. and Min.*, vol. 10, 1899

nations, appear to have ceased by degrees, and the circulating solutions became, therefore, principally aqueous solutions which, at relatively low temperature, strove to effect among the earlier formed vein materials the re-combinations possible under these changed conditions. The products of mineral deposition in cavities still open (or produced by destructive work), as well as the products of pseudomorphosis of the earlier formed vein minerals, must, therefore, now become different; observation shows that almost exclusively waterbearing silicates—zeolites—were thereby formed. Just as little as we could draw a sharp boundary between the first and second phases, can we perceive the second and third phases of vein formation to be sharply differentiated from each other. Indeed, the formation of sulphurbearing ores continued after the beginning of zeolite formation. . . .

That the zeolites have been formed at relatively low (although not ordinary) temperature has been determined with certainty through the researches of Daubrée, Lemberg, de Schulten and others; apparently the successive changes of temperature with constant decrease thereof has influenced the formation of zeolites in this manner, that certain zeolites can form preferably within a somewhat higher, others within a somewhat lower range of temperature. It appears that in that way most simply can the sequence of the zeolites be explained, which everywhere is in the main nearly constant in the veins of the border-zone.

The sequence for the deposition of zeolites in open vugs is as a rule the following:

Analcite.	
Eudidymite,	Thomsonite.
	Stilbite,
Apophyllite.	

Of all the zeolites formed in open vugs, analcite is the oldest and also the most widespread in our veins. . . .

The analcite is often formed in greatest extent at the expense of the elaeolite; in part, however, other soda-rich silicates, especially sodalite and albite, have also given rise to the formation of analcite, as will shortly be mentioned below. Where it has been formed from elaeolite, so much substance appears to have been removed simultaneously in part with its appearance that apparently new open druses were thereby formed, in which besides the analcite other minerals also could later be deposited. In part, however, the analcite has been deposited in older, preëxisting vugs, for it rests often on crystals of leucophane and fluorspar, for example, of the second phase of vein-formation. . . .

The next zeolite in the sequence is eudidymite, which is only known from one vein upon the Island of Ober-Ärb. It rests here upon analcite and is itself covered with natrolite. . . .

The normal sequence of zeolites in the vugs of our veins shows natrolite after analcite. . . .

Apparently simultaneously or nearly simultaneously with the formation of natrolite, that of thomsonite began.

In a single specimen belonging to the Imperial Museum from an unknown occurrence of our veins, fine blades of stilbite rest upon thomsonite needles as

a younger formation in secondary vugs in a hydronephelite spreustein formed from elæolite.

Younger than the natrolite is apophyllite in all occurrences where it appears; it is, however, in comparison with the soda-rich zeolites—*analcite* and *natrolite*—almost always present in wholly subordinate amount. I have not observed it with *stilbite*; it is, therefore, not possible to say with certainty whether the *stilbite* or the *apophyllite* is the older of the two; but to judge by the character of its occurrence, the last may be considered with some probability as almost always the youngest of all zeolites of our veins. *Apophyllite* also belongs to the zeolites of *Plombières* made known by *Daubrée*, formed at a late period at only about 70°.

4. Phase of fluo-carbonates and carbonates, etc.

... As the youngest formation in the vugs, perhaps deposited at almost ordinary temperature, one finds here and there calcite, usually only compact, seldom in idiomorphic crystals—for example, upon *natrolite* of *Risö*; only traces of a chloritic (?) mineral have I observed as a still younger deposit upon the calcite.

RÉSUMÉ

The zeolites and associated secondary minerals of the Watchung basalt are found in certain localized areas of the sheet. These areas possess well-marked peculiarities of structure which are plainly of primary origin and due to conditions present at the time of consolidation. The chief peculiarity in these limited areas is an accumulation of boulder-like masses of "pahoehoe" lava, which consist of an interior of normally crystallized basalt and an exterior crust of glass. From a study of the geological relations in the field, it is believed that the basalt sheet represents a surface flow of lava poured out over a continental area which had been depressed by crustal movements of deformation (either warping or faulting) and in whose lowerlying portions a lake or series of lakes occupied shallow basins. The pahoehoe structure of the basalt is believed to have been developed over or immediately adjacent to the lake beds through quicker cooling of the flow.

The secondary minerals are held to have been developed from the elements of the basalt (especially the glassy crusts) and from sublimates given off by the magma in cooling, and deposited in crevices of the flow. The medium in which recrystallization took place was an aqueous solution, which was enabled to percolate through the sheet of basalt, because of the more permeable structure above the lake beds. A purely meteoric origin is indicated for the waters, which probably worked in from the edge of the sheet through the porous sandstones below and returned to

the surface through the interstices of the basalt in the situations described.

Recrystallization was probably begun as soon as the lava had cooled sufficiently to permit the waters to hold a liquid form. A considerable degree of superheating was permitted because of the head to which they were subject. By degrees, however, the whole mass of lava became cooler, so that in the final stages of alteration, the temperature may have been but little above the average climatic temperature of the region.

The majority of the secondary minerals are found to consist of the elements present in the original plagioclase feldspar, with or without the addition of water. The possibilities of equilibrium among the great number of compounds present was considered in the light of Gibbs's phase rule, and some inquiry was made into the applicability of this rule to the conditions of the problem. It was deemed most probable that, although various modifying conditions entered whose effect could not be wholly taken into account, the general conclusions to which the phase rule pointed should hold and that the possibility of all the compounds present coexisting in equilibrium could not be admitted.

Petrographic examination of thin sections confirmed these conclusions, and the sequence of minerals and the form taken by processes of replacement have been described at length. It was found that in addition to the zeolitic minerals which form such a prominent feature of the occurrence, and to such minerals as prehnite, datolite, quartz etc., which are present in notable amount, traces of other minerals have survived. Such are albite, garnet, arfvedsonite etc., which represent the effects of intense metamorphism in the early stages of recrystallization. There is reason to believe that these were formed in considerable quantities, but were almost destroyed during succeeding stages.

It was found possible to trace very perfectly the alteration of the glass, or less often the normal basalt, into various associations of secondary minerals; and several features present in the original rock were found to have left traces through all stages of recrystallization. In some instances, the theory of solid solutions was of assistance in affording a basis of explanation of the changes which minerals had undergone. This was especially true in the case of the development of albite from original labradorite and in the transformations of the amphiboles.

As a final effect of alteration, quantities of calcite were formed. The greater part of this is believed to have been produced at temperatures but little above the ordinary. The presence of the carbonic acid is attributed to a derivation from the atmosphere at the beginning of the cycle of circulation.

Comparison was made between the Watchung deposit and similar mineral formations elsewhere. In some cases, there was evidence that conditions during the formation of zeolites were different, or that the subsequent history had effected changes which made impossible a strict comparison with the Watchung rocks. In most cases, however, judging from the rather scanty evidence available, conclusions regarding sequence are confirmed.

PLATE XI

MAGNIFIED SECTIONS OF BASALT AND SECONDARY MINERALS

- FIG. 1. Normal crystallization of dense basalt. $\times 24$. Slide 18.
- FIG. 2. Dense basalt bordered by secondary albite. $\times 24$. Slide 62.
- FIG. 3. Dense basalt bordered by secondary albite (Ab) with faultlike groups of prehnite (Pr) and some calcite (Ca). $\times 24$. Slide 62.
- FIG. 4. Groups of radiating quartz crystals surviving from an early period of alteration in a slide in which zeolites and chlorite form the principal features. $\times 24$. Slide 59.
- FIG. 5. Garnet (Gr), fibrous green amphibole (Am) and prehnite (Pr) encrusting dense basalt. The garnet forms clear grains where it borders the prehnite, but the greater portion is in masses which are nearly opaque from dust of TiO_2 or MnO_2 . The circular markings are interpreted as due to nuclei of resorbed olivine in the original glass which the secondary minerals have replaced. $\times 24$. Slide 53.
- FIG. 6. Garnet grains (Gr), acicular amphibole (Am) and long needles of pectolite (Pe) in prehnite (Pr). $\times 24$. Slide 50a.



PLATE XII

MAGNIFIED SECTIONS SHOWING ALTERATION IN BASALT

- FIG. 1. Breccia of an original glass, whose interstices have been filled with ferruginous clay from the lake bottom. Recrystallization of the glass has resulted chiefly in albite and chlorite. $\times 24$. Slide 12.
- FIG. 2. Incipient stage of zeolitic alteration of glass, showing effect of chill cracks in directing percolation of solutions. $\times 24$. Slide 30.
- FIG. 3. Alteration effects in brecciated glass. $\times 24$. Slide 61.
- FIG. 4. Prominent banding in secondary products, due to cracks in the original glass. The darker portions are chiefly chlorite, the lighter analcite. $\times 24$. Slide 60.
- FIG. 5. Geometrical patterns in garnet, interpreted as due to resorption of olivine. Gr = garnet, Am = amphibole, Pr = prehnite. $\times 32$. Slide 115.
- FIG. 6. Incipient alteration of glass, showing accentuation of physical and chemical differences by a slight degree of alteration. $\times 35$. Slide 54.

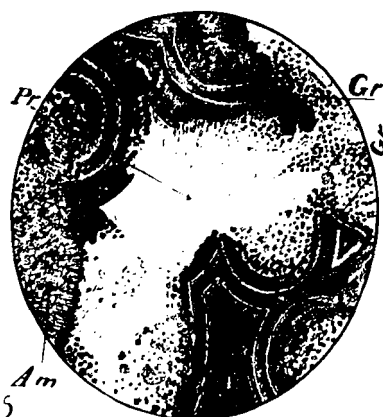
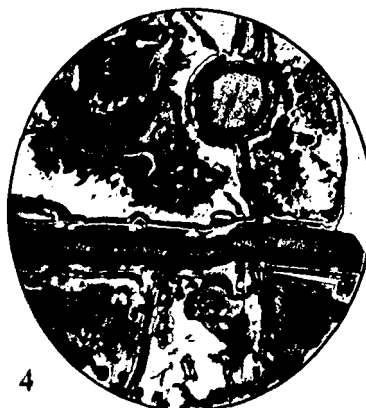


PLATE XIII

MAGNIFIED SECTIONS OF SECONDARY MINERALS IN BASALT

- FIG. 1. Association of garnet (Gr), fibrous green amphibole (Am), albite (Ab), prehnite (Pr), specularite (Sp), chabazite (Ch) and calcite (Ca), forming a crust on dense basalt. $\times 35$. Slide 98.
- FIG. 2. Association of garnet, amphibole and prehnite. $\times 35$. Slide 109.
- FIG. 3. Complex forms of secondary minerals found in an advanced stage of alteration of glass. The darker portions are chiefly glass which has suffered little alteration. Rounded remnants of several diopside crystals appear, and a number of small altered plagioclase laths. The clear bands and vermicular lines are zeolitic alteration products, largely analcite. $\times 38$. Slide 3.

