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It is surely worth while for us to pause a moment in this time of rapid advance of all scientific knowledge to consider the present and future state of mineralogical science as compared with that of a generation ago. Such a comparison should serve to bring out the chief advances which have been made and may even suggest some features of the progress that may be expected in the future.

Thirty years ago students of rock components were interested primarily in the surfaces and average composition of minerals; today they are studying the internal structure and variations of composition of minerals.

The study of the surfaces of crystallized minerals developed into the science of crystallography and permitted keen thinkers to reach certain correct conclusions regarding the internal structures of crystals, but these conclusions did not attain general acceptance because of lack of definite proof. Our knowledge of crystals resembled that which might be obtained of houses and public buildings, entirely devoid of windows and doors, by a study confined entirely to their exterior surfaces. Thirty years ago the average composition of minerals was determined by chemical analysis, but this gave no information regarding the distribution of the constituents in the crystal and resembled the information one might obtain concerning a public building or palace by completely wrecking it and then measuring the total weight of all the brick, all the stone, all the mortar, all the steel, and all the lumber used in its construction. Nothing would be known regarding the manner of use of the materials in the construction of internal and external walls and the arrangement of rooms and galleries in the original building.

Today the new x-ray methods have enabled us to study the internal structures of crystals and to learn the precise arrangement

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and nature of the constituent parts. The new knowledge of crystals resembles that which would be obtained by making a careful study of all the internal features of a public building or palace which had previously been accessible only to external observation and to study after complete demolition. The mineralogist who had hitherto been barred from entrance has now found an open door into his crystal palace and is using it to learn the whole story of the internal arrangements and nature of walls, rooms and galleries.



FIG. 1. Variations in composition and optic properties in the camsellite-sussexite series.

Thirty years ago the mineralogist considered that each mineral had some definite and simple composition corresponding with some simple chemical formula. Today he has learned that such a condition is very rare in nature and that nearly all minerals vary considerably in composition. Many minerals may vary in composition continuously from one chemical formula to a different one. Such variations do not change the essential nature of the mineral, although its properties show gradual variations corresponding with the gradual changes in composition. Also, a single mineral may vary in composition in more than one way.

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An example of the simplest kind of variation in composition is furnished by the series from camsellite, $HMgBO_3$, to sussexite, $HMnBO_3$. Such a series is conveniently represented by a line divided into 100 parts so that the percentage composition represented by any point may be read off promptly; also, the variations in properties may be expressed easily by means of a suitable scale on the second coordinate, as illustrated in Fig. 1.

One substance may vary in this way to more than one other substance. For instance, siderite, $FeCO_3$, may vary to magnesite,



FIG. 2. Variations in composition and optic properties in the glaucochroitemonticellite-CaFeSiO₄ system. Based on data of Beliankin and Ivanov: Am. Jour. Sci., XXII, 1931, p. 72.

 $MgCO_3$; or to rhodochrosite, $MnCO_3$; magnesite apparently does not vary directly to rhodochrosite. The main facts regarding such a double series can be shown by merely doubling the length of the horizontal line used to show the variations in composition and still using the other coordinate to express variations in physical properties. This is illustrated by a diagram¹ showing the relations be-

¹ A. N. Winchell, Am. Mineral., XVII, 1932, p. 473.

tween variations in composition and in properties in three kinds of tourmaline.

In some compounds one element may be replaced by any combination of two other elements in any proportions and then all variations in composition can not be represented by a line, but may be shown by a triangle. This is approximately illustrated by the condition in the CaFeSiO₄-CaMgSiO₄-CaMnSiO₄ system exhibited in Fig. 2, which shows by means of lines similar to contours the variations in optical properties corresponding with the variations in composition.

In certain compounds one element may be replaced by any combination of three other elements in any proportions; for such cases a solid tetrahedron is needed to show all possible variations in composition. Fortunately, illustrations of this condition seem to be rare in natural minerals so far as known at present.

In case a mineral varies in composition in more than one way, one element in the mineral may be replaced by some other element, and also a different element of the original mineral may be replaced quite independently of the first replacement. For example, in diopside, CaMgSi₂O₆, the Mg atom may be replaced by Fe, and thus diopside may vary to hedenbergite, CaFeSi₂O₆, and, wholly independent of this change, the Ca atom may be replaced by Mg and thus diopside may vary also to clinoenstatite, MgMgSi₂O₆. If diopside varies in both these ways the result is obviously clinohypersthene, MgFeSi₂O₆. All the possible variations in such a system may be shown conveniently on a square, as in Fig. 3, one corner of which represents pure diopside, another corner pure hedenbergite, a third corner pure clinoenstatite, and the fourth corner pure clinohypersthene. Then, if each side of the square is divided into one hundred parts, these divisions will show the percentage composition represented by any point. Thus, in Fig. 3 the point A on the base line represents 57% of hedenbergite and 43% of diopside, the point B on the left hand side represents 39% of clinoenstatite and 61% of diopside. Similarly a point C inside the square represents 29% of the Mg-Fe change (or 29% hedenbergite+clinohypersthene) and 42% of the Ca-Mg change (or 42% clinoenstatite+clinohyperstheme); therefore it represents 29% of 42% or 12.2% of clinohyperstheme. In an entirely similar way the percentage composition represented by any point in the square in terms of the four end-member molecules may be obtained readily from the figure.

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The relations between optic properties and chemical composition in this system are shown in Fig. 4. Known examples of clinohypersthene are so rare that the optic properties of that substance



FIG. 3. Variations in composition in the clinoenstenite-diopside-hedenbergite system.

are obtained partly by extrapolation from the properties of the three other end-members of the system.

Thus far, this discussion has dealt only with variations in composition which can be expressed by varying an atom in the formula of a mineral, a procedure which corresponds with replacing (more

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or less completely) all the atoms of a certain kind in the crystal structure by atoms of a different kind. Now it has been discovered that some kinds of crystals can change their composition in another way, namely, by losing some of the atoms they normally possess, or by taking on atoms not present in the crystal as first



FIG. 4. Variations in composition and optic properties in the clinoenstenitediopside-hedenbergite system.

formed, or by both these changes. Some changes of this kind lead to substances which can not be produced directly. Let us consider, first, some examples of changes involving loss of constituents. Sulphuric acid attacks biotite in such a way as to dissolve out all the bases and leave merely scales of silica, still possessing the approximate structure and optical properties of a mica. No way is known to produce such siliceous scales directly. Again, certain zeolites² can be deprived of all their constituents, except SiO₂, (by treatment with HCl) and the remaining silica (especially after heating) has optical properties closely related to those of the original zeolite. No way is known to produce such zeolite structures from pure silica directly. Once again, brucite, $Mg(OH)_2$, when properly heated, loses its water, but still retains the approximate structure and optic properties of the original mineral, instead of changing to periclase. No way is known to produce such a structure directly from magnesia. Or again, the hydrogen can be driven out of ferriferous amphiboles³ (with oxidation of the iron) without destruction of the crystal structure, although the optic properties change considerably in this case. It seems probable that such anhydrous amphiboles can not form directly from their components.

Next we may consider examples of changes involving the addition of some constituents; such changes are not common probably because most crystallized minerals are close-packed aggregates of atoms. However, magnetite, Fe_3O_4 or Fe_6O_8 , may take on additional oxygen so as to become Fe_6O_9 (or Fe_2O_3) without any important change in its crystal structure, although Fe_2O_3 crystallizes directly to an entirely different structure. Again, the ferrous iron in chlorite seems to be oxidized readily to the ferric state with no considerable change in the structure or properties of the mineral. It seems unlikely that such chlorite can be produced directly from ferric iron oxide and the other necessary constituents.

Third, there are some minerals which may lose some normal constituents and take on others which are not normal. For example, many zeolites may be heated so as to drive out the water without destroying the crystal structure; upon cooling in the absence of water vapor they will absorb alcohol, or ammonia, or CO_2 , or CS_2 , or glycerine, or vapors of mercury, or bromine, or iodine, or merely air, while still retaining essentially the same crystal structure. It is probable that such zeolites have never been formed directly. Also, each calcium atom of a natural zeolite may be replaced by two sodium atoms, or by two silver atoms, or by two copper atoms, or by two thallium atoms; while each pair of sodium atoms of a natural zeolite may be replaced by one atom of calcium, or barium, or strontium. Examples of such zeolites are not unknown in nature,

² F. Rinne: Crystals and the Fine Structure of Matter, 1924, p. 243-157.

³ V. E. Barnes: Am. Mineral., XV, 1930, p. 393.

but it seems probable that they were not formed directly. Such exchanges are not confined to zeolites; they are important in soils, and they supply much of the mineral matter needed for plant growth. They are also well shown by other hydrous minerals, as illustrated by calcium uranyl phosphate, called autunite, in which the calcium may be replaced under valence control by sodium, potassium, barium, manganese, copper, lead, or magnesium. Also, the phosphoric acid in this mineral may be easily replaced by vanadic acid, or arsenic acid, apparently without destroying the crystal. It is probable that some of these products of substitution can not form directly.

A very important feature of such changes is that they lead in some cases to products that can not be formed directly; therefore, whenever such products are found in nature, it is possible to reach certain conclusions regarding the history of the rocks in which they are found. For example, zeolites abnormally rich in soda imply that the rocks in which they occur have probably been affected by alkaline solutions since the formation of the zeolites. Chlorites abnormally rich in ferric iron imply that the rocks in which they are found have probably been permeated by hot oxidizing solutions since the formation of the chlorites. Amphiboles deprived of hydrogen and thus containing an abnormal tenor of ferric iron imply that the rocks in which they are found have probably been heated above 800°C. since the formation of the amphiboles. Unfortunately such conclusions are only probabilities at the present time; much more complete knowledge of all the conditions of formation of such products is needed to make similar conclusions certainties.

An illustration of the uncertainties which befog the situation at present is afforded by the facts now available as to the formation of oxydized magnetite or oxymagnite. No way is now known to make oxymagnite crystals directly from Fe₂O₃, but it is possible to make a mixture of various proportions (up to about 50% Fe₂O₃) of spinel and Fe₂O₃ crystallize with the oxymagnite crystal structure. The variations in composition and optic properties in such a series are shown in Fig. 5, as deduced from the experimental work of Fisk and McCaughey.⁴

These variations have been described and illustrated at some length chiefly to emphasize the main point, which is, that minerals

⁴ Ohio Univ. Stud., Eng. Ser. I, July, 1932.

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commonly vary considerably in composition and properties. These variations are indeed so common and so great that mineralogists are no longer agreed as to what constitutes a mineral. Many of the mineral names in common use seem to apply most accurately to end-members of a series, or a system, which cannot be divided into parts except by purely artificial rules or limits. This condition is doubtless a direct result of the former view that a mineral had a definite composition corresponding with a simple chemical formula. That view has had other consequences also, some of which are quite



FIG. 5. Variations in composition and refractive index in the spinel-oxymagnite series. Based on data of Fish and McCaughey: *Ohio Univ. Stud., Eng. Ser. I*, July, 1932, page 28.

unfortunate. For example, it is generally recognized that minerals vary in their physical properties. Since they were supposed to be constant in composition, it was formerly held that minerals varied in optical properties in spite of fixed composition, and therefore that accurate measures of optical constants were needless, because of no significance in naming the mineral. It is only very recently and very gradually becoming apparent that any fixed and definite composition within the limits of variation of any mineral has fixed and definite physical characters. This correlation of physical and chemical properties was impossible in many cases as long as the min-

eralogist had no key to unlock the door of his crystal palace. Now that he has entered these palaces he has learned for the first time to understand how they vary in composition; such a knowledge was necessary in order to make it possible to show the relations between physical characters and chemical composition. Such a knowledge was also necessary in order to enable us to discriminate between variations in composition which are possible at the time of formation of the crystal and those which are only possible after the formation of the crystal palace.

The study of minerals in the old way (merely in terms of names) may be compared with the study of animals or plants in terms of isolated species; a mineral (in the proper meaning of the term) corresponds in a certain sense with a genus rather than with a species.

The aim in the scientific studies of minerals should no longer be merely the name of the mineral, but the determination of the precise composition in terms of end-members and, for this purpose, accurate measures of optical constants are second in importance only to complete chemical analyses of carefully purified samples.

By recognizing post-formational changes of composition the mineralogist of the future will have a new key to unlock the secrets of the past.