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METHODS AND INSTRUMENTS USED IN MINERALOGY*

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Examination of the materials at and near the surface of the earth proves that, if we disregard plants and animals, they consist of various kinds of masses, such as rocks, soils, and ores of different types, disposed according to definite patterns expressive of their modes of formation and subsequent history. Closer study reveals that these rocks and ores consist in turn of one or more substances of different properties and aspects. These smaller units are called minerals. Each mineral is more or less homogeneous in chemical composition and is commonly recognized wherever it occurs by certain distinguishing characteristics or features.

Mineralogy is the branch of science which describes and classifies minerals, ascertains the physical and chemical properties of each mineral and determines its crystal form and crystal structure, if it is crystallized.

Interest in minerals and crystals extends back to primitive peoples who were attracted to them by the variety of forms and colors which they present and by their appeal when properly cut and mounted in jewelry. The perfection of workmanship and the art and skill attained by these peoples in the shaping and mounting of various minerals and gems is extraordinary. Their tools and methods were of the crudest type; but perseverance and the urge to make ornaments for themselves and for votive offerings to their gods accomplished results that delight the archaeologist and give pleasure to us of the present day. Intrinsically beautiful, they serve as important historical documents indicative of the state of culture of the races that fashioned them.

It is the object of this talk to consider briefly a few of the methods and instruments now in use by mineralogists in determining the physical, crystallographical, and chemical properties of minerals. Experience has proved that in experimental work the introduction of a new method or a new instrument may enable the observer to explore new fields heretofore inaccessible because of lack of the proper weapons of attack. Under present war-time surroundings this condition is forcibly impressed upon us

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and we realize the importance of adequate supplies of the instruments of warfare and of new methods and weapons of defense and of offense.

In 1609 Galileo made a telescope by grinding and polishing a simple biconvex objective lens and a biconcave eyepiece lens and mounting them at the opposite ends of a tube. With this new optical instrument, of which he had had only a brief description from Holland, he was able to make ships at a distance appear much closer. The quality of the glass in his lens elements was poor and, at the magnification of 33 diameters obtained by his telescope, the quality of the image was extremely poor. However, on January 9, 1610, he pointed his telescope at the heavens, at the sun, the moon, Venus, and Jupiter, and began a series of exciting discoveries. He studied the movement of sun spots and proved that the sun is rotating; he studied the moon and found, that, contrary to the opinion of philosophers, its surface is not smooth but mountainous in certain areas; he measured roughly the heights of several lunar mountains by the lengths of the shadows they cast; he studied Venus and noted its phases not unlike those of the moon; he studied Jupiter and discovered its four large satellites, recorded their periods of revolution and concluded that he had before his eyes a miniature system similar to that postulated by Copernicus for the solar system. It has been said that Galileo's observations on the satellites of Jupiter served to convince people of the validity of the Copernican theory of the solar system in spite of the opposition to the theory by the Church, and in later years to Galileo himself. The introduction by Galileo of the telescope to the study of the heavenly bodies opened a new era in astronomy.

Forty years later the compound microscope was invented in Holland. By its use much of interest, especially in the biological field, was discovered; but the quality of images produced by it was too poor to permit adequate magnification. For 150 years its usefulness was restricted because of poor design of the objective lens and poor quality of available glass. The result was that this tool now so successfully employed in mineralogical work did not at once give to mineralogy the impetus that the telescope gave to astronomy. However, in 1669 Nicolas Steno did observe the shapes of crystals precipitated from solution on a slip of glass and asserted that, for a given chemical salt, the shapes of the crystal faces formed may vary from crystal to crystal but not the interfacial angles. This is the first statement of the constancy of crystal angles more definitely proved a century later by Romé de l'Isle in 1783 and by Haüy in 1784 with the aid of the contact goniometer invented by Carangeot, assistant to Romé de l'Isle.

Reflecting goniometer. The contact goniometer was followed in 1809 by the single-circle reflecting goniometer of Wollaston. With its introduction

precision measurement of crystal angles began and engaged the attention of crystallographers in Europe. In 1874 Miller constructed a two-circle goniometer by adding a vertical circle to the usual horizontal circle instrument; this arrangement was modified by Fedorov in 1889 who used an autocollimating telescope in place of the collimator and telescope. In 1893 three two-circle goniometers were described, one by Fedorov, the second by Czapski, and the third by Victor Goldschmidt. Of these the design by Goldschmidt has proved to be the best and is now widely used and has been of the greatest aid to crystallography. In 1889 and 1904 G. F. Herbert Smith described two models of a three-circle goniometer. Impressed by the usefulness of the measurement of crystal faces in a zone, he employed the third circle to aid in the setting of a crystal so that the angles in any one of its zones can be measured directly on the horizontal circle. The three-circle goniometer was also used for the solution of spherical triangles. Although the three-circle goniometer is interesting and serves its purpose well, it has not been adopted as a tool by mineralogists.

Crystal goniometers have served for purposes other than the measurement of angles. In 1889 V. Goldschmidt described a cutting goniometer for making models of actual crystals. In 1894 Tutton exhibited his single-circle crystal-grinding goniometer which he used later in preparation of the crystal plates and prisms for his extended investigations into the isomorphous series of the orthorhombic sulphates and selenates of potassium, rubidium, and cesium, and into other isomorphous series. In 1915 F. E. Wright described a two-circle crystal-grinding goniometer for grinding faces of any desired orientation on a crystal and accurate in position to one-half minute of arc. Orientated faces on crystals of quartz, calcite, and other minerals were ground with the aid of this apparatus and used in the measurement of the changes of crystal angles with changes of temperature. For the measurement of crystal angles at elevated temperatures a special electric resistance furnace, water-cooled on the outside and mounted on the horizontal plate of the Goldschmidt goniometer was constructed and measurements were made to temperatures of 1250° C. For measurements at low temperatures V. M. Goldschmidt constructed a special apparatus for surrounding the mounted crystal with an atmosphere approaching in temperature that of solid carbon dioxide, or of liquid air.

The equipment at present available for the measurement of crystal angles is satisfactory and adequate for crystals of appreciable size; but there is still room for improvement in the measurement of minute crystals and of small crystal faces only one-tenth millimeter in width. The amount of light reflected from such faces is so small that the reflection signals are

weak and spread out as a result of diffraction. At present the best procedure appears to be the use of an intense carbon arc lamp as a source of illumination together with a telescope having an objective lens of short focal length and nice adjustment of the entire optical system. The best results will be obtained with a goniometer of special design.

Instrument design. In this connection it is well to state that each instrument intended for precision work should be designed with reference both to purpose and rugged mechanical performance and to adjustment facilities, so that the observer will be in a position at any time to test the accuracy of adjustment of all parts of the instrument by an orderly procedure of successive adjustments so arranged that no later adjustment disturbs preceding adjustments. If this precaution is not taken, the observer is forced either to devise a makeshift method of adjustment, which is never satisfactory, or to place implicit faith in the skill of the manufacturer to furnish the instrument in correct adjustment and to rely upon maintenance of the adjustment in spite of the jars and jolts of transportation and the inevitable slight movements upon aging of the metals of which the instruments are made. Another result of long and sometimes bitter experience is the futility of the so-called universal instruments of a generation ago. Instruments of this type are likely to be unwieldy and of relatively poor design, so that they are not satisfactory for any one of the several kinds of measurement for which they are intended.

Projections. In the study of the crystallographical and optical properties of crystals certain projections are extremely useful. They enable the observer to gain oversight of the spatial relations under investigation and to obtain approximate values of the relative positions and distribution of the important points and directions in space. Experience has shown that different projections are needed for different purposes.

In crystallographic measurements the gnomonic projection (ascribed to Thales, 650 B.C.) is especially useful for plotting the polar and azimuthal angles of measurement; also for preparation of crystal drawings. In this projection crystal zones are represented by straight lines. The limitation of the projection is its large distortion and the fact that polar angles in excess of 70° are not convenient to plot.

The stereographic projection (Hipparchus, 150 B.C.), which is angle-true (conformal, orthomorphic) and in which all zones are great circles, is useful both for crystallographical and for optical work. It covers the hemisphere easily, but distorts polar angles increasingly as the margin of the hemisphere is approached. Another perspective projection, the orthographic (Hipparchus), in which the hemisphere to be projected is viewed from an infinite distance, is used but little in crystallographical and optical work. Interference figures, as observed in the petrographic micro-

scope, are orthographic projections of the interference phenomena observed in convergent polarized light and their measurements are made as in this projection.

Several other projections, the Lowry perspective and the globular or equidistant, in polar and meridional forms, distort less than the preceding projections and have this advantage in crystal optical studies. The equidistant projection is not a mathematical projection in the sense that each point of the sphere is projected according to a definite mathematical formula, but rather as a mode of representing the sphere on a plane by a series of easily constructed curves.

In recent years students in the field of petrofabric analysis, or study of the internal structure of a rock, have made extensive use of the Lambert equal-area meridional projection net, described by Lambert in 1772; this projection has been employed in cartographic work and atlases continuously since its introduction by Lambert. In 1925 Schmidt applied this projection net to problems in petrofabric analysis and in that field the projection is called the Schmidt projection net. It would seem that this practice of renaming a long established projection net, or in fact any other well known appliance or tool for accomplishing a given purpose, is not justified nor in keeping with scientific accuracy. A case similar to the 'Schmidt' net is the 'Wulff' stereographic net in use in crystal optics. Fortunately this designation is but little employed at present and the name stereographic projection net serves the purpose adequately. In the case of the Lambert equal-area projection net, however, the designation Lambert is needed to distinguish it from other equal-area projections in common use in cartographic work.

The petrographic microscope. For the measurement of the optical properties of minerals the petrographic microscope is the most important instrument available to mineralogists. Its development did not follow immediately upon the introduction of the reflecting goniometer. During the years 1814 to 1818 Sir David Brewster wrote a series of papers announcing his discovery of biaxial crystals and of the intimate relations between optical and crystallographical relations in crystals. In 1828 William Nicol, the inventor of the nicol prism, ground thin sections of rocks and mounted them in Canada balsam. In 1850 H. Clifton Sorby studied thin sections of rocks; between 1850 and 1861 he published the results of his investigations of certain rocks and demonstrated the possibilities of the petrographic microscope in the study of thin sections of rocks. He was followed in 1863 by F. E. Zirkel, who described a series of rocks. In 1870 and 1876, H. Rosenbusch constructed a practical petrographic microscope with rotating stage. During that decade interest in the petrographic microscope became general. Such microscopes were

manufactured in several countries and some of them are still in use. At the present time a number of petrographic microscopes of satisfactory design and quality are on the market.

The most important optical constants of a mineral for various wave lengths of light are: its principal refractive indices; its principal birefringences; the nature of the optical ellipsoid of reference (index ellipsoid); determination of the spatial relations between the principal planes of its index ellipsoid and its principal crystallographic directions (extinction angles for definite directions and zones).

Refractive indices are measured most easily by the immersion of fine mineral grains in refractive liquids or solids of known refractivity for a given wave length of light at a definite temperature. A number of liquids and mixtures of liquids are available to cover the range in refractive indices between 1.33 and 2.10; and in low melting solids to cover refractive indices up to 3.0 and even in excess of 3.0. For the comparison of the refractive index of a fine mineral grain with that of the surrounding liquid, either the Becke-line method or that of oblique illumination is used. The degree of accuracy attainable by these methods, under carefully controlled conditions, is about 2 in the fourth decimal place; at this limit the difference in intensity of illumination between the Becke-line, or oblique illumination effect, and the general field is just detectable by the eye; below it the gradational contrast is no longer perceptible to the eye. Determinations accurate to ± 0.001 in refractive index are easily made on favorable grains measuring only a few microns in diameter. For measurements in different parts of the spectrum, either the liquid mixture must be changed or the same liquid held at different temperatures. The decrease in refractive index of a liquid is commonly 0.001 per 2° to 3° C. rise in temperature. Determinations of the refractive indices of liquids are made either with the aid of a total reflecting refractometer or by the prism method on a spectrometer or goniometer. In either case only a drop of the liquid is required.

Birefringence measurements are made on crystal plates of uniform, known thickness by use of a birefracting graduated wedge or by tilting a plane-parallel plate to compensate the path difference produced by the mineral plate. In this measurement the thickness of the mineral plate is the chief element of uncertainty. Optic axial angles are determined by imaging the interference figure, formed in convergent polarized light, on a finely divided coordinate scale in the focal plane of a positive eyepiece; the scale may also be placed in the lower focal plane of the substage condenser and may then be graduated to read off angular distances directly. The angular equivalent of each scale division is ascertained either by computation or preferably by direct determination with the aid of an

Abbe apertometer or of the collimator of a goniometer. The optical character of a birefracting mineral is ascertained by use of a birefracting plate or wedge of known orientation.

Experience has shown that it is possible to determine these optical properties, and other less important optical characteristics, on mineral grains measuring only one one-hundredth millimeter in diameter.

Thin sections of rocks and of mineral aggregates are used primarily for the study of the morphological development of the various minerals and the relations between the mineral grains, rather than for the accurate measurement of their optical properties. With the Fedorov stage, however, determinations of optic axial angles and of positions of the planes of optical symmetry relative to crystallographic directions, especially in the feldspars, are made on thin sections of rocks. In recent years the Fedorov stage has been employed with great success in petrofabric analysis, to ascertain for a given mineral, such as quartz, the relative frequency of occurrence or distribution in space of a given crystallographic direction, such as the principal or optic axis. From this study of grain orientation in a rock mass, by use of sections cut along definitely known planes in a rock specimen whose orientation in the field has been ascertained, information is gathered on the present structure of the rock mass and on its deformation in the past. This combination of field and laboratory study is directly dependent on the information yielded by measurements with the Fedorov stage which in this field of scientific inquiry is indispensable.

Crystal structure. In 1784 Abbé René Just Haüy discovered the law of rational intercepts for crystals, stated the laws of crystal symmetry, showed that all varieties of crystal forms can be referred to a few types of symmetry, and that, for a given crystal substance, its cleavage and other crystal faces are related to a simple primitive form, characteristic of the crystal. He proposed an hypothesis for the orderly spatial arrangement of the 'molecules integrantes' within a crystal which finds expression in flat crystal faces and in the rationality of their intercepts on three crystallographical axes of reference. Although Haüy's explanation of crystal structure emphasized the idea that crystal units are stacked in parallel orientation, it was proved later to be defective for physical reasons; but it did arouse interest in crystals and their structure. This interest has continued to the present time.

Following Haüy, both crystallographers and mathematicians contributed to the purely geometrical aspects of the problem. By the end of the last century the problem had been solved through the labors chiefly of Hessel, Bravais, Gadolin, C. Jordan, Sohncke, Curie, Schoenflies, Fedorov, Barlow, and others. These studies were based on the assump-

tion that matter is not continuous but granular in nature and consists of atoms which in these studies may be considered to be indivisible and to be located at very small, but not indefinitely small, distances apart. In a solid these atoms may be clustered into chemical molecules, or groups of molecules (crystal particles), which in turn serve as its building units. In a crystal these units are in orderly arrangement, such that the field surrounding any single unit is exactly similar to that surrounding any other unit, with the exception of units situated near boundary surfaces. The crystal is thus homogeneous and each crystal particle, or fundamental cell, has the same orientation in space, while within each crystal particle the arrangement of the component atoms and molecules (groups of atoms) is exactly the same as in each other crystal particle. These assumptions lead to the conceptions of arrangements of the crystal particles along definite directions in space, to the grouping of these lines in definite planes, and to spatial groupings of parallel planes which constitute spatial crystal lattices. The study of the symmetry relations within these crystal lattices, the types of movements of translation and of rotation through finite distances to produce self coincidence led first to the establishment, by Bravais in 1848, of fourteen possible space lattices in which the crystal is considered to consist of molecules, alike and similarly oriented, whose centers of mass occupy the points of the space lattice. The Bravais fourteen space lattices encompass all holohedral crystals. Additional lattice types were developed by Sohncke through consideration of the positions of the atoms within the crystal particles and the resulting series of interpenetrating lattices. These considerations raised the number of possible crystal lattices to sixty-five. If the principle of enantiomorphous symmetry is introduced, the total number of geometrically possible structures on the assumption of three independent translations and no infinitesimal translation becomes two hundred and thirty; this number was reached independently by three investigators, Fedorov, Schoenflies, and Barlow, and by quite different methods of approach. It was furthermore proved that there are thirty-two, and only thirty-two, finite groups of movements that are consistent with the law of rational indices and hence valid in crystallography. These constitute the basis for the thirty-two classes of crystal symmetry groups.

In addition to the solution of the geometrical problems presented by space lattices possible in crystals, crystallographers had made great progress in the study of the behavior of isomorphous series of crystals and also of the effects produced on the crystallographical and physical properties of solutions in the crystal state by the step by step substitution of one metal base by another, and had learned much regarding the spatial relations within the crystal particle itself. They had also learned to think

in terms of face normals rather than in actual crystal faces and in crystal indices rather than in axial intercepts; also in polar forms and their graphical expression in the gnomonic projection. The time had arrived for the introduction of a new tool that would enable the investigator to probe into the actual crystal unit cells, to ascertain the relative positions of its component atoms, and to measure the exact distances between them.

This new tool was discovered by Roentgen in 1895 and named by him *x*-rays. He found that *x*-rays penetrate ordinary materials with ease. The next two decades were occupied by physicists in exploring the nature of *x*-rays and in putting them to practical uses, especially in the medical field.

In 1912 Laue, interested in *x*-rays and acquainted with the concepts of space lattices as developed by crystallographers, suggested that the space lattice of a crystal might serve as a space grating for *x*-rays and produce diffraction patterns on a photographic plate, if they were of the nature of light waves and of wave lengths smaller than the minute spacing intervals of a crystal space lattice; in other words, of lengths of the order of one ten-thousandth of the wave lengths of light waves. The experiment was tried by Friedrich and Knipping by sending an *x*-ray beam through a thin plate of copper vitriol. The result on the photographic plate was a diffraction pattern of adequate clarity to indicate the correctness of Laue's theoretical conclusion. The interest aroused by this experiment and others on crystals of the cubic system spread quickly; in the following year W. H. Bragg and his son, W. L. Bragg, introduced the spectrometer ionization-chamber reflection method which was widely adopted and served both in the analysis of the crystal structure of many crystal types and in the measurement of *x*-ray wave lengths themselves.

In 1916 Debye and Scherrer, and independently A. W. Hull, introduced the powder method which enabled observers to analyze crystal structures by the photographic method on crystal powders. The method was also adapted for use with the ionization chamber and spectrometer.

Still another method was developed in which a crystal of known orientation is rotated at constant speed about an axis normal to the impinging *x*-ray beam and coincident with the axis of a cylinder which is stationary and which carries a photographic film mounted against its inner surface. The resulting negative shows a series of diffraction spots spaced along parallel layer lines; by measurement both of the separation of the diffraction spots along the layer-lines and of the distances between the layer-lines, the cylindrical coordinates of each diffraction spot can be determined and, except for an element of uncertainty regarding identification of certain spots, the space lattice of the crystal can be determined.

This element of uncertainty was removed in 1924 by Weissenberg by use of the x-ray goniometer in which, during rotation of the crystal as in the simple rotation method, the photographic film is moved at a definite rate of translation and without rotation. A heavy metal cylinder with a narrow slot aperture cut normal to its axis is placed between the rotating crystal and the film in such position that only diffracted beams from a single layer-line are permitted to impinge on the film. At least four negatives are taken, one from the equatorial layer-line, one from the first layer-line, another from the second, and so on. This procedure is called the normal beam technique and is more rapid in application than the rigorous Bragg spectrometer ionization-chamber reflection technique. These two methods are, however, complementary; the Bragg method is particularly well adapted to the measurement of large crystals; the Weissenberg method to that of small crystals.

Recently an important advance has been made by M. J. Buerger by his equi-inclination beam method in which the axis of the cylinder and of the rotating crystal of the Weissenberg x-ray goniometer bisects, for a given layer-line photograph, the angle between the x-ray beam and the reflected ray. Buerger showed that the equi-inclination projection used in connection with photographs taken by this method has an advantage over the normal beam procedure because it requires only a single template for all curve forms and provides in addition straight lines for central lattice lines. The equi-inclination method was also shown by George Tunell to possess advantages in respect to the trigonometric functions involved in the intensity computations.

Thus far we have considered only three tools useful to mineralogy: the reflecting goniometer, the petrographic microscope, and x-ray apparatus. The field is too vast to do more than this in a short paper. The effort has been made rather to illustrate how progress in mineralogy has been dependent on the availability of suitable methods and instruments of attack; also that an instrument useful in one branch of science may prove to be equally serviceable in another field, if properly adapted to meet the conditions imposed in the new field. No attempt has been made to predict the applications of new methods, such as electron diffraction, or of new instruments, such as the electron microscope, to mineralogical problems. These must be gradually tested to ascertain their possibilities and their limitations before their usefulness in the field of mineralogy can be stated. We may rest assured, however, that mineralogy will continue to progress and that many discoveries important to mineralogy and to human welfare will be made by the mineralogist interested in research work in science.