

FLUID INCLUSIONS IN BERYL AND QUARTZ FROM
PEGMATITES OF THE MIDDLETOWN
DISTRICT, CONNECTICUT

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ABSTRACT

Secondary and apparently primary fluid inclusions occurring in quartz and beryl from certain pegmatites of the Middletown district, Connecticut, have been investigated by the Sorby method. In three pegmatites the inclusions found appear to be aqueous solutions of moderate concentration. In the other pegmatites most or perhaps all the inclusions contain considerable amounts of carbon dioxide. Temperature data for quartz have proved difficult to obtain, owing to fracturing and leakage of inclusions when the crystals are heated. Beryl has yielded considerable data. The data are analyzed in terms of possible physico-chemical conditions in pegmatitic liquids and in terms of the various sources of error inherent in the Sorby method. The bearing of the work on the use of liquid inclusions in determining temperatures of pegmatite mineral formation and compositions of pegmatitic liquids is discussed.

INTRODUCTION

Since 1938, field studies of pegmatites and of the occurrence of minerals in them have directed attention to a series of problems that call for laboratory studies correlated with the field investigations. The need for these studies became apparent to the first-named author during wartime investigations of pegmatite mineral deposits, and a group of beryl-bearing pegmatites in the Middletown district of Connecticut was selected for study. These pegmatites had been mapped in detail for the U. S. Geological Survey by Cameron and V. E. Shainin during the period 1942-44. In the fall of 1945, Cameron made further studies of the pegmatites in the field and collected suites of specimens. Laboratory study was undertaken at the University of Wisconsin in the fall of 1949.

The present paper is a report of the first phase of the study, during which fluid inclusions in beryl and quartz from various pegmatites have been investigated. The chief objects were to learn whether the fluid inclusion method can be used to determine temperatures of mineral formation in this group of pegmatites, and to obtain a check on temperature data of formation reported for liquid inclusions in beryl and quartz from two of the pegmatites. The investigation has also yielded information that may bear on the composition and physical characteristics of the fluids from which pegmatites crystallize.

ACKNOWLEDGMENTS

The laboratory studies have been made possible by funds granted by the Wisconsin Alumni Research Foundation. Weis participated in the

project from September 1949 to June 1950, and Rowe participated from September 1950 to June 1951. The support of the Foundation is gratefully acknowledged. We are also greatly indebted to several members of the faculty of the University of Wisconsin for assistance and advice, in particular to Professor J. R. Dillinger, of the Department of Physics, and to Professor C. H. Sorum, of the Department of Chemistry.

PREVIOUS WORK

Certain pegmatites of the Middletown district have been known for many years as sources of commercial feldspar and mica, and as prolific sources of mineral specimens. There are numerous articles dealing with the pegmatites; a few are especially pertinent to the present investigation.

Jenks (1935) described the results of a detailed mineralogical study of the Strickland-Cramer pegmatite. During 1942-45, many of the pegmatite bodies of the district were mapped in detail and studied by E. N. Cameron and V. E. Shainin; these studies are described in a forthcoming report. A brief description of the more important beryl-bearing pegmatites has been given by Cameron and Shainin (1947).

In 1947, Ingerson gave temperature data for liquid inclusions in beryl and quartz in specimens furnished by Cameron from the Case No. 1 and No. 2 pegmatites. Ingerson reported that bubbles in inclusions in several sections of quartz and one of beryl disappeared at temperatures ranging from 153° C. to 175 °C. Quartz from one of the specimens studied by Ingerson was subjected to decrepitation by H. S. Scott (1948, p. 650), who obtained 165° C. as the temperature of formation of the quartz (uncorrected for pressure).

DESCRIPTION OF THE PEGMATITES

Location and geologic setting: The pegmatites presently being studied are mostly in a narrow strip east of the Connecticut River and parallel to it, in the vicinity of Middletown (Fig. 1). They are essentially lenticular bodies of various sizes enclosed in the Bolton schist and the Monson (Glastonbury) gneiss. One body, the Slocum pegmatite, lies east of the others, in the town of East Hampton. It occurs in interlayered schists and schistose quartzites known as the Hebron gneiss (Gregory and Robinson, 1907). The metamorphic rocks of the area are involved in a series of major folds. Age determinations from radioactive minerals obtained from several pegmatites of the district suggest that the pegmatites are of late Devonian age (Foye and Lane, 1934), and the enclosing rocks are generally considered to be of Paleozoic age.

There is no direct evidence of the thickness of rocks that has been re-

moved since emplacement of the pegmatites. Indirect evidence suggests that the thickness may have been great. The area forms part of the Eastern Highland of Connecticut. The Triassic rocks of the Central Connecticut Lowland rest on an erosion surface that presumably extended eastward and truncated the folded, crystalline rocks of the Eastern Highland. Erosion of the Eastern Highland contributed much of the 10,000 to 15,000 feet of sediments comprising the bulk of the Triassic formations,

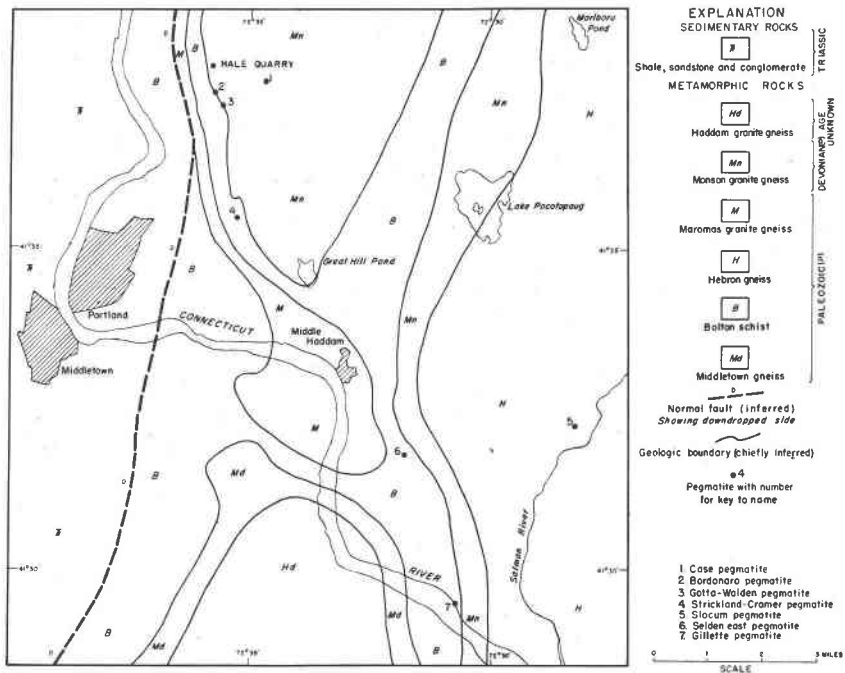


FIG. 1. Index map of the Middletown district, Connecticut, showing locations of pegmatites studied.

but how much the surface developed on the crystalline rocks was lowered during that time is a matter of conjecture. An erosion surface correlated by D. W. Johnson (1931) with the Schooley peneplane truncates both the Triassic rocks and the rocks of the Eastern Highland. Furthermore, if Johnson's interpretation of the geomorphic history is correct, this surface truncates the earlier Fall Zone peneplane, on which the coastal plain sediments of Long Island rest. The history of the region since the time of folding and metamorphism therefore includes two and possibly three periods of prolonged erosion. An estimate of 15,000 feet for the minimum thickness of covering rocks removed since emplacement of the pegmatites

would therefore seem conservative. The true thickness may be considerably greater.

Character of the pegmatite bodies: The pegmatite bodies of the area range from those that are essentially uniform in mineral composition and mineral proportions to those in which zonal structures are developed in various degrees. The pegmatites that form the basis of the present investigation are a selected group, consisting largely of bodies in which zonal structures are at least moderately well developed. The sole exception is the Selden east pegmatite, in which an indistinct wall zone and pods of quartz rimmed by coarse feldspar and muscovite are the only indications of zonal structure.

The pegmatites are mostly simple in composition. The essential minerals are perthite, sodic plagioclase, quartz, and muscovite or biotite, or both. Garnet, tourmaline, and beryl are accessory minerals in all. The many other minerals found from time to time in the pegmatites are trace components.

The zones and corresponding mineral assemblages of the pegmatites are summarized in Table 1.

TABLE 1. ZONES AND MINERAL ASSEMBLAGES OF THE PEGMATITES

Pegmatite	Border zone	Wall zone	Intermediate zone		Core or apparent core	Replacement bodies
Case No. 1	<i>P, Pl, Q, B</i>				<i>P, Q, B, G, M</i>	<i>Pl, Q</i>
Case No. 2	<i>P, Pl, Q, M, G, B, T</i>	<i>P, Pl, Q, M, B</i>	<i>P, Q, B</i>		<i>Q</i>	
Case No. 3	<i>P, Pl, Q, M, G, B, T</i>	<i>P, Pl, Q, M, B, T</i>	<i>P, Q, Pl, B</i>		<i>Q</i> (pods)	
Bordonaro south	<i>Pl, P, Q, B, M</i>	<i>P, Q, M, Pl</i>			<i>Q</i>	
Bordonaro north	<i>Pl, P, Q, M, B</i>	<i>Pl, Q, P, M, B, T, G</i>			<i>P, Q, Pl, M, B, G</i>	<i>Q, Pl</i>
Gotta-Walden	<i>Q, Pl, M, G, T, B</i>	<i>Pl, M, Q, B, G, T</i>			<i>Pl, Q, P, M, B, G, T</i>	
Selden east	<i>Pl, Q, M, B</i>	<i>Pl, Q, M, P, B, G, T</i>			<i>Q, Pl, P, M, B</i>	
Strickland-Cramer	<i>Q, M, Pl, G, T</i>	<i>Pl, Q, M, T, G</i>	(1) <i>Q, P, Pl, M, G, T</i>	(2) <i>Q, Pl, G, T</i>	<i>Q, Pl</i>	<i>Pl, Q, B, M, Li minerals</i>
Slocum	<i>Q, P, Pl, G, T, B</i>	<i>Q, P, Pl, B, T, M, Bi</i>	<i>P, Q, B</i>		<i>Q, P</i>	

Symbols: P, perthite; Pl, plagioclase; Q, quartz; M, muscovite; Bi, biotite; G, garnet; B, beryl; T, tourmaline. Symbols for essential minerals italicized. Trace minerals omitted.

OCCURRENCE OF BERYL AND QUARTZ IN THE PEGMATITES

Beryl is unevenly distributed in most of the pegmatites. It is abundant in certain zones or replacement bodies but rare or absent in others. In the Strickland pegmatite, for example, visible beryl is largely restricted to replacement bodies, whereas in the Case No. 2 and Case No. 3 pegmatites it is concentrated in the border and wall zones. In the Gotta-Walden pegmatite, however, beryl is found in approximately equal amounts in all three zones of the body.

Beryl occurs in all pegmatites as subhedral to euhedral prismatic crystals. The crystals are 6 inches or less in length and 2 inches or less in diameter in most of the pegmatites, but larger crystals have been found in a few. In inner zones of the pegmatites, the crystals are commonly haphazardly oriented, but in outer zones beryl generally forms crystals oriented perpendicular or subperpendicular to the walls. These crystals are commonly tapered. The outer ends (nearest contacts of pegmatite with wall rock) are slender, whereas the inner ends (toward the cores of the pegmatites) are thicker. In addition, the beryl crystals in some of the pegmatites are progressively larger from zone to zone from the walls inward.

Beryl varies in color from pegmatite to pegmatite or even within the same pegmatite. Beryl from the Bordonaro, Gotta-Walden and Case pegmatites is medium green. Beryl from the Selden east pegmatite is pale green, nearly white. Beryl collected at the Slocum mine is golden, although beryl of other colors is said to have been found there. Beryl from the Strickland-Cramer mine ranges from blue to golden to pale pink. The color variations presumably correspond to variations in alkali content.

Quartz occurs in all zones and replacement bodies of all the pegmatites. It occurs mostly as anhedral grains and aggregates of anhedral grains that are interstitial to other minerals. It ranges from clear to milky to smoky in color.

APPARATUS USED FOR TEMPERATURE DETERMINATIONS

A. Apparatus for heating experiments

The apparatus used for heating experiments and the procedure for preparation of crystal plates are essentially the same as that described by Bailey and Cameron (1951, p. 634). For high-temperature work, however, a cell of smaller diameter was found more suitable. It has also been found best to polish only the upper side of the crystal plate. The lower side is ground with M-303 (American Optical Co.) emery. This ground surface provides a diffuse illumination against which inclusions can be seen more clearly through the polished upper side of the plate.

In practice, the polished plate is examined carefully under the microscope before heating runs are attempted. Liquid inclusions present are studied and classified into groups according to shape, distribution, and arrangement with respect to crystallographic features. Each group is studied separately. The approximate liquid-vapor ratios shown by the inclusions are noted. A series of inclusions is then selected for heating trials. Sketches are made showing the individual inclusions in each field of view that will be watched during heating. The inclusions are numbered, so that effects of heating can be recorded separately for each inclusion.

Heating is done at a controlled rate, in order to prevent a lag of the temperature of the plate with respect to that of the cell. It has been found that a rate of 10–15° C. per minute is satisfactory. The inclusions are watched throughout the heating process. Temperatures of vapor disappearance or of phase boundary disappearance are read by means of a potentiometer and thermocouple. For actual reading, the tip of the thermocouple is touched to the lower side of the crystal plate. The potentiometers used are calibrated against the melting points of standard alloys. Experience indicates the temperature values given in the present paper for temperatures of disappearance of a vapor phase or of a liquid phase are accurate to within 2½° C. Values for disappearance of phase boundaries are less accurate and may be as much as 10° C. in error; the precise point at which disappearance is complete is difficult to determine.

The end result of heating most fluid inclusions is disappearance of the vapor phase. When this has been accomplished, heating is discontinued and the cell allowed to cool. The temperature of reappearance of the vapor phase is observed as a rough check on the temperature of disappearance. After cooling, each run is repeated as a check on the first determination and as a check against leakage.

Disappearance of the liquid phase or of a phase boundary is similarly checked by repeated measurements.

Many of the inclusions examined contain a second liquid phase that disappears when heated to a temperature close to 31° C. We infer this to be liquid carbon dioxide. This second liquid phase is easily overlooked, because the heat of a strong lamp may raise the temperature of inclusions above 31° C. As a check, the plate must be cooled by moistening it with acetone or alcohol and watching the results under the microscope.

Various combinations of 10× and 20× hyperplane objectives with 10× achromatic, 20× achromatic, and Leitz UM 4 objectives have been used in the present study. The UM 4 objective combines large working distance with high magnification (30×) and is especially useful for study of minute inclusions. A water-cooled sleeve device serves to prevent heating of the objective when in use.

B. Apparatus for freezing experiments

Results of heating experiments raised various questions as to the composition of the liquid inclusions studied. Freezing experiments have therefore been undertaken. A simple wooden cell has been used for this purpose. Solid carbon dioxide or a current of air passed over liquid air contained in a thermos jar has been used as the coolant. With liquid air, temperatures as low as -120° C. have been reached, but condensation of moisture in the cell has caused trouble, and a dehydration device is needed.

Subzero temperatures are read by means of a thermocouple connected to a Leeds and Northrup factory-calibrated potentiometer. The temperature within the cell can be controlled within 1 or 2 degrees C. by regulating the flow of air over the liquid air.

RESULTS OF THE STUDY OF FLUID INCLUSIONS

General statement: It was intended originally to study fluid inclusions in quartz from all nine pegmatites listed in Table 1, but it soon became apparent that quartz in these pegmatites is ill suited to temperature studies. One reason is that primary and secondary inclusions in quartz are difficult to distinguish. Most of the liquid inclusions that are not clearly secondary occupy irregular cavities. The grains are anhedral and unzoned, hence the distribution of inclusions with relation to crystallographic elements is difficult or impossible to determine. The second reason is that quartz shows a marked tendency to fracture when heated, and most liquid inclusions leak before temperatures of disappearance of the vapor phase are reached. Reliable temperature determinations for inclusions in quartz are therefore very difficult to obtain. Beryl, on the other hand, occurs in well-formed crystals. The distribution of inclusions with respect to crystallographic elements is in general easily determined. Many inclusions occupy oriented negative crystal cavities or tubular cavities that show no relation to secondary structures of the crystals, and there is a strong possibility that at least some of these can ultimately be shown to be primary. Further, the shapes of many inclusions are such that during heating the changes in the volumes of the component phases can be determined accurately. Finally, the crystals show little tendency to fracture, and leakage of inclusions is not a serious problem; the majority of inclusions can be heated to temperatures 50° C. or more above the filling temperatures. Beryl is therefore very satisfactory for temperature studies and has received our principal attention.

Fluid inclusions in beryl: More than 2,000 fluid inclusions in beryl from the various pegmatites have been studied in the course of the pres-

ent investigation. Temperatures of disappearance of the vapor phase, the liquid phase, or the vapor-liquid phase boundary have been determined for some hundreds of fluid inclusions. The majority of the determinations are precise measurements, inclusion by inclusion. In certain beryl crystals from the Selden, Case No. 2, and Strickland-Cramer pegmatites, however, clusters of inclusions could be observed simultaneously during heating. Precise disappearance temperatures sufficient to represent the range for each cluster have been made, and the temperatures of disappearance for the remainder have been determined approximately.

Fluid inclusions found in crystals of beryl are of 3 principal types:

- (1) Disseminated negative crystals and tubular inclusions with long axes parallel to the c -axis of beryl (Figs. 2 and 3). These are referred to below as oriented negative crystals. In an earlier paper (Cameron, Rowe, and Weis, 1951) the tubular inclusions were regarded as a separate type, but experience has shown that a sharp line between tubular inclusions and negative crystals can-

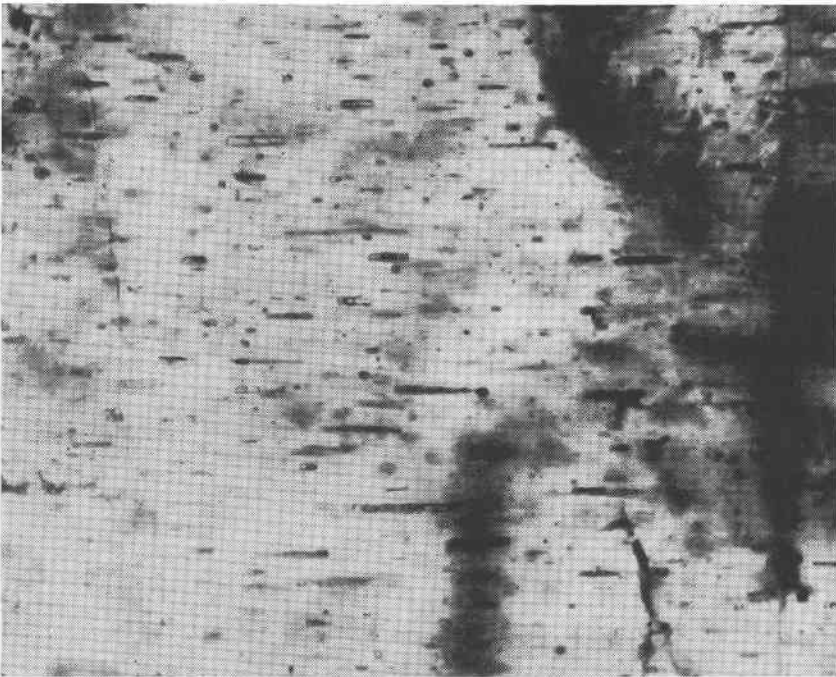


FIG. 2. Negative crystal and tubular inclusions in beryl, oriented parallel to the c axis. Case No. 2 pegmatite. $\times 200$.

- not always be drawn. Both may occur in the same mineral plate.
- (2) Disseminated rounded, elongate, or irregular inclusions, unoriented with respect to crystallographic elements (Fig. 4).
 - (3) Rounded, elongate, or irregular inclusions and negative crystals arranged along planes or curving surfaces of random orientation.



FIG. 3. Negative crystal inclusions in beryl, oriented parallel to the c axis. Selden east pegmatite. $\times 200$.

Intersections of planes or surfaces are common (Fig. 5). In most of the plates cut these are the only inclusions. In most of the remaining plates, they are the most abundant type of inclusion.

Inclusions of type (3) are definitely secondary. They are distributed along healed fractures unrelated to crystal boundaries or crystallographic elements of the host beryl. The origin of the other types of inclusions is less certain. Those of type (2) show no relation either to crystallographic elements or to secondary structures. They may be either primary or secondary. Inclusions of type (1), on the other hand, are crystallographically oriented and appear unrelated to fractures or other discontinuities within the crystals. Inclusions of this type evenly disseminated in swarms in the centers of unfractured crystals are especially difficult to account for as secondary inclusions. They were clearly developed prior to the inclusions of type (3), which have a pattern that is superimposed upon the pattern of inclusions of type (1). In this connection, a large crystal of golden beryl from a replacement body in the

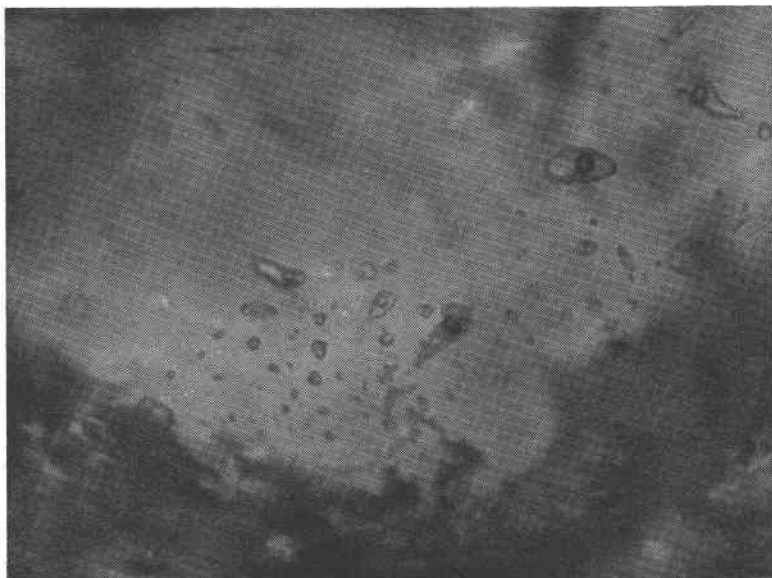


FIG. 4. Disseminated irregular inclusions in beryl, Strickland-Cramer pegmatite. $\times 200$.

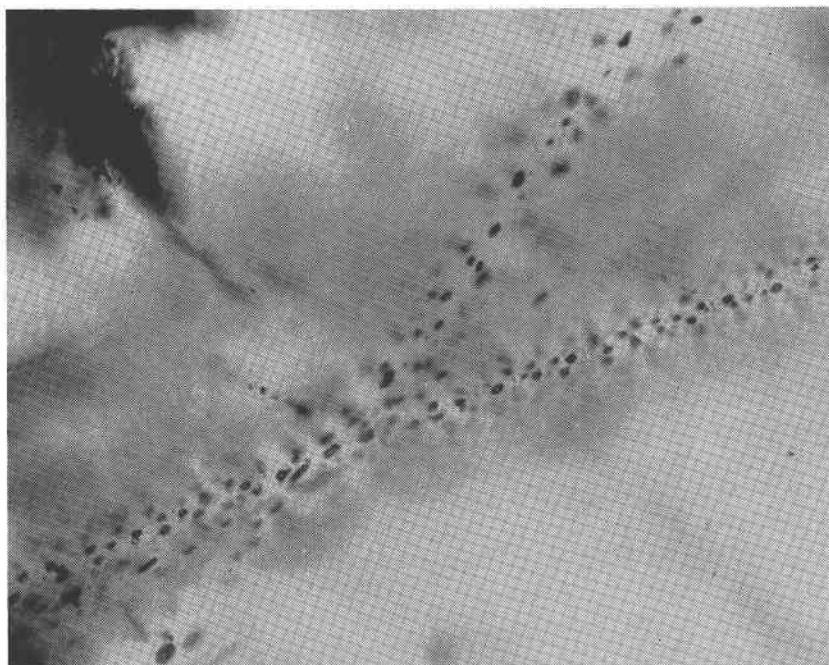


FIG. 5. Secondary inclusions along a branching healed fracture in beryl, Bordonaro north pegmatite. $\times 100$.

Strickland-Cramer pegmatite is worth noting. This crystal is veined along its basal parting by quartz and cleavelandite deposited later in the development of the replacement body. The veins are several inches apart. Plates cut from this crystal show that the distribution of inclusions of type (1) in the beryl is unrelated to the veins. The inclusions occur in swarms away from the veins as well as adjacent to them. If the inclusions formed during or after the development of the veins, it seems extraordinary that they show no relation to so obvious a discontinuity.

As yet we find no absolute proof that inclusions of type (1) are primary, but it seems probable that material selected with specific reference to the problem of origin will show that inclusions of this type developed during beryl formation. For the present, but solely for convenient reference and distinction from the obviously secondary inclusions, we shall refer to them as the "apparently primary inclusions."

These inclusions are divisible into two main groups,* according to their behavior when heated. One group is represented by inclusions discovered in beryl from the Selden east pegmatite (Cameron, Rowe and Weis, 1951 p. 908) and subsequently recognized in beryl from the Strickland-Cramer pegmatite. At room temperature, each inclusion consists of a vapor phase plus a liquid phase. Heating causes the vapor phase to shrink and finally to disappear. Temperatures of disappearance (precise determinations only) are given in Fig. 6. For the Strickland-Cramer pegmatite, all but two of the precise temperature values shown in the figure fall within the range 210° C.-260° C. Approximate values for a large number of additional inclusions fall within the same range. For beryl from the Selden east pegmatite, values obtained range from 187° C. to 386° C.; most of the precise values plotted, together with approximate values for about 150 additional inclusions not shown in Fig. 6, are between 210° C. and 290° C. Nothing in the crystals themselves suggests an explanation of the range of values. Some inclusions less than a tenth of a millimeter apart give markedly different values of the temperature of disappearance. On the other hand, certain clusters of inclusions give values remarkably close to one another. The values obtained have no relation either to the sizes of inclusions or their positions within crystals.

The fluids composing inclusions of this type are of high mobility even at room temperatures, and their composition is of much interest. Fortunately, some of the tubular inclusions are highly elongate and of uniform diameter, hence the ratio of the length of the part of the cavity occupied by liquid at a given temperature to the total length of the

* The grouping of inclusions in the present paper differs from that employed by the authors in a brief note in this journal, vol. 36, pp. 906-910. Additional work has shown that Group B recognized in that paper belongs under the second main group described below.

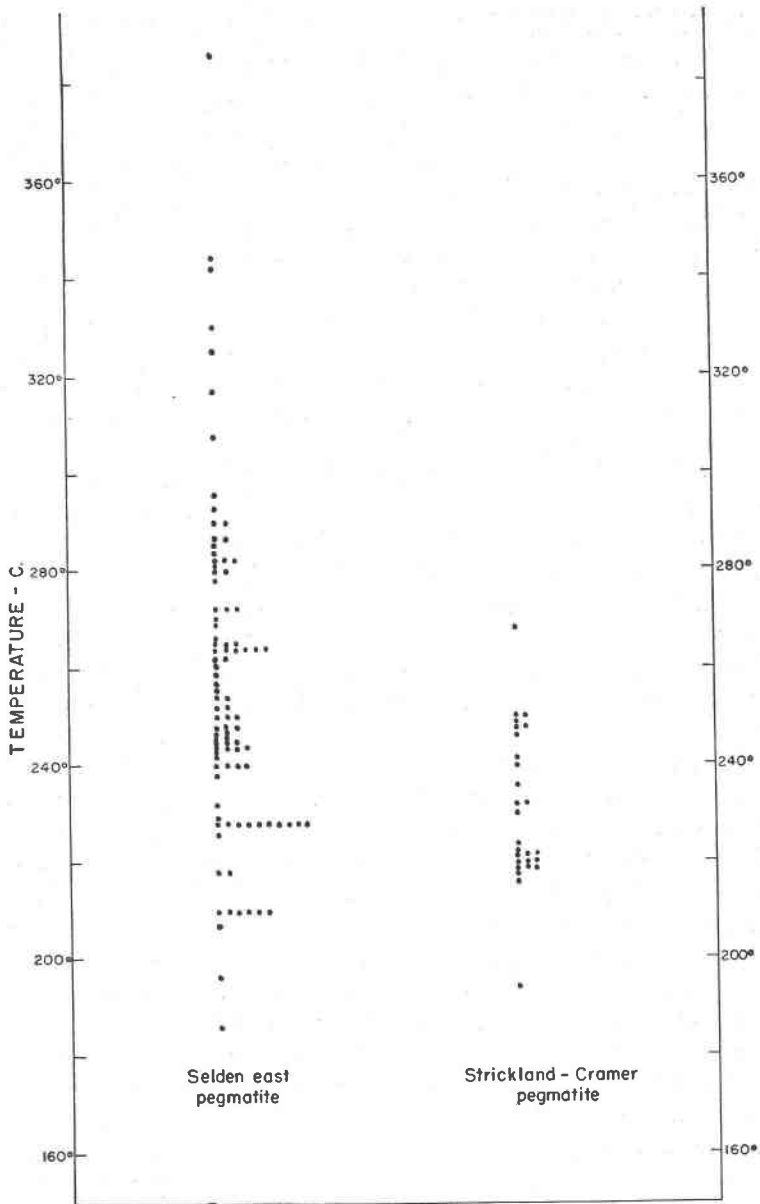


FIG. 6. Temperatures of disappearance of the vapor phase in two-phase apparently primary inclusions in beryl from the Selden east and Strickland-Cramer pegmatites. Each dot represents one inclusion.

cavity should closely approximate the ratio of volume of liquid to volume of cavity. Changes in this ratio with heating have been determined by Rowe for some of the tubular inclusions, and curves for a representative pair of these are shown in Fig. 7. With the curve for each inclusion a curve is also given for a hypothetical inclusion composed of pure water that would exactly fill its cavity at the temperature of filling of the actual inclusion. In both cases the two curves are very close to one another, and this suggests that the inclusions either approach pure water or are composed of some fluid remarkably similar to water in thermal characteristics. Furthermore, the melting points of the phase formed by freezing the liquid in 5 inclusions of this group in beryl from the Selden pegmatites range from -5° C. to -12° C. These data likewise suggest aqueous solutions. Inasmuch as the possible salts can only be guessed, the concentrations of the solutions can be estimated only roughly, but recorded data on melting points of solutions of various salts in water point to concentrations in the range between 2.7 and 6.5 molar.

The second group of apparently primary inclusions is considerably different from the first. At room temperature, these inclusions show three phases,—two liquid phases plus a vapor phase (Fig. 8, *A, C*) At about 31° C., the boundary between the vapor phase and one liquid phase fades and disappears, so that the inclusions consist only of the vapor phase plus a single liquid phase (Fig. 8, *B, D*). As 31° C. is the critical temperature of carbon dioxide, we infer that the disappearing phase is essentially liquid carbon dioxide. Owing to difficulties with our present cooling cell, we have not yet been able to determine the melting point of the phase.

All apparently primary inclusions observed in beryl from the Case No. 1 and Case No. 2 pegmatites, the Bordonaro north and south pegmatites, the Gotta-Walden pegmatite, and the Slocum pegmatite are of the three-phase type at room temperature. All those observed in beryl from the Case No. 2 pegmatite, and some of the inclusions in beryl from each of the other four pegmatites, show shrinkage and final disappearance of the vapor phase on heating. Temperature data (precise determinations only) for these inclusions are shown in Fig. 9. The ranges of values are as follows:

<i>Pegmatite</i>	<i>Number of inclusions</i>	<i>Temperatures of disappearance of the vapor phase</i>
Bordonaro north	23	282° – 353° C.
Bordonaro south	2	299° – 338° C.
Case No. 2	30	292° – 332° C.
Slocum	4	310° – 455° C.
Gotta-Walden	6	238° – 355° C.

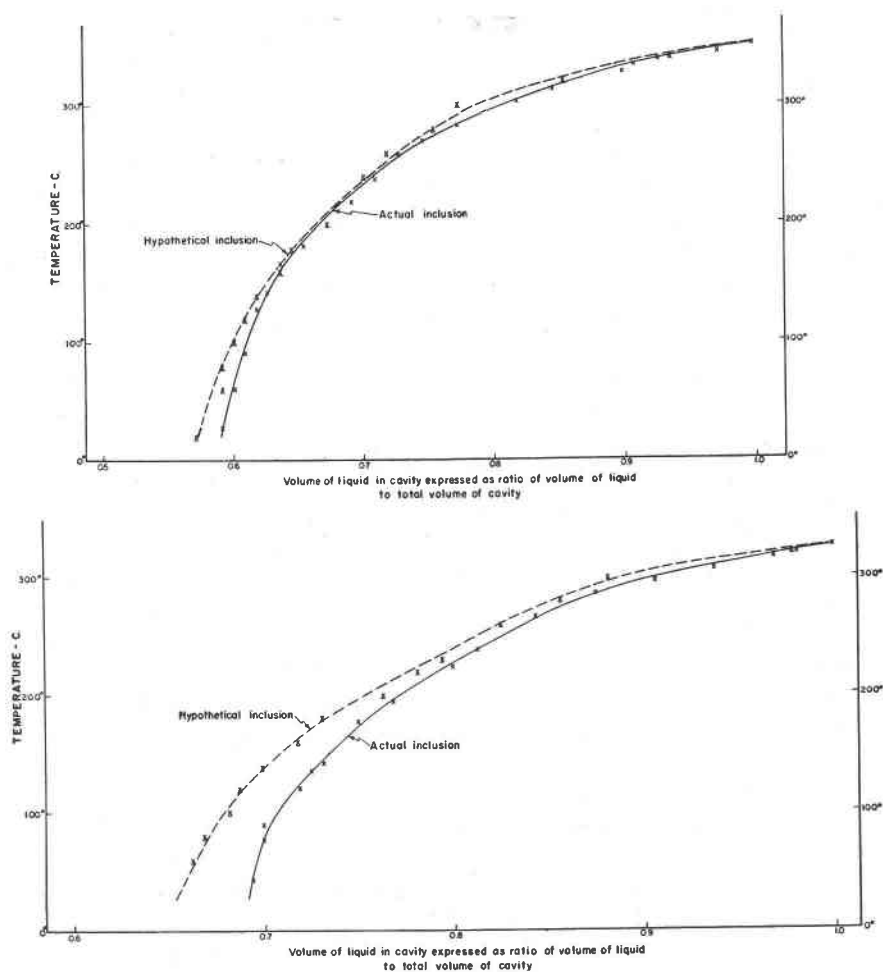


FIG. 7. Curves showing changes in volume for actual two-phase inclusions in beryl from Selden east pegmatite and for hypothetical inclusions composed of pure water. In each case, both the actual inclusion and hypothetical inclusion are filled with liquid at the temperature corresponding to the upper end of the pair of curves.

The changes in volume of liquid when an inclusion of this type is heated have been determined for 6 inclusions. In Fig. 10, the curves for two such inclusions are plotted against the comparable curves for a hypothetical inclusion composed of pure water that would be filled at the same temperatures as the actual inclusions. The curves for the three-phase inclusions differ markedly from those for the water inclusions, and furthermore lie on the opposite side of the curves for the water

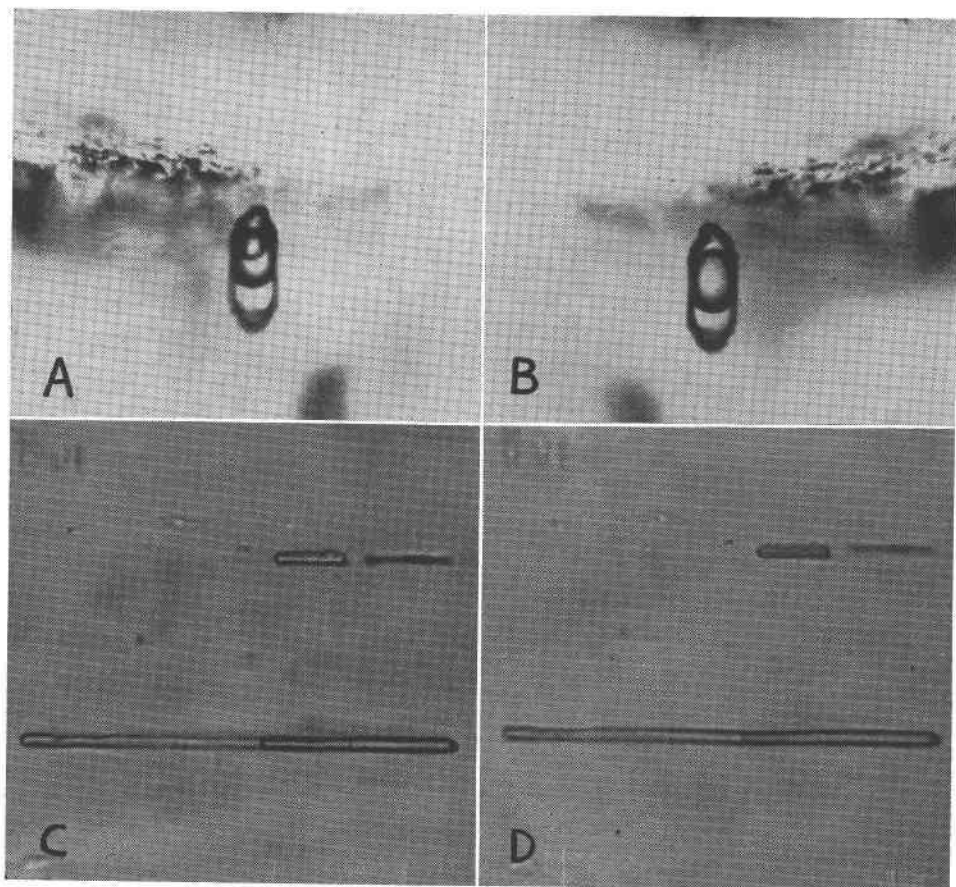


FIG. 8. Three-phase inclusions in beryl. (A) Oriented negative crystal inclusion in beryl, Bordonaro north pegmatite, at room temperature, showing 3 phases. $\times 200$. (B) Same inclusion, heated above 31° C. (C) Oriented tubular inclusion in beryl, Case No. 2 pegmatite, at room temperature, showing 3 phases. $\times 460$. (D) Same inclusion, heated above 31° .

inclusion from the curves for the two-phase inclusion of Fig. 6. The presence of significant amounts of carbon dioxide in the three-phase inclusion is presumably responsible for the marked difference.

Many inclusions of the three-phase type, when heated, behave differently from those described. According to their precise behavior, they fall into three subgroups. In the first, the vapor phase expands until it occupies the entire cavity. Inclusions having this behavior occur in two beryl crystals from the Bordonaro north pegmatite, in one crystal from the Bordonaro south pegmatite, in one crystal from the Case No. 1

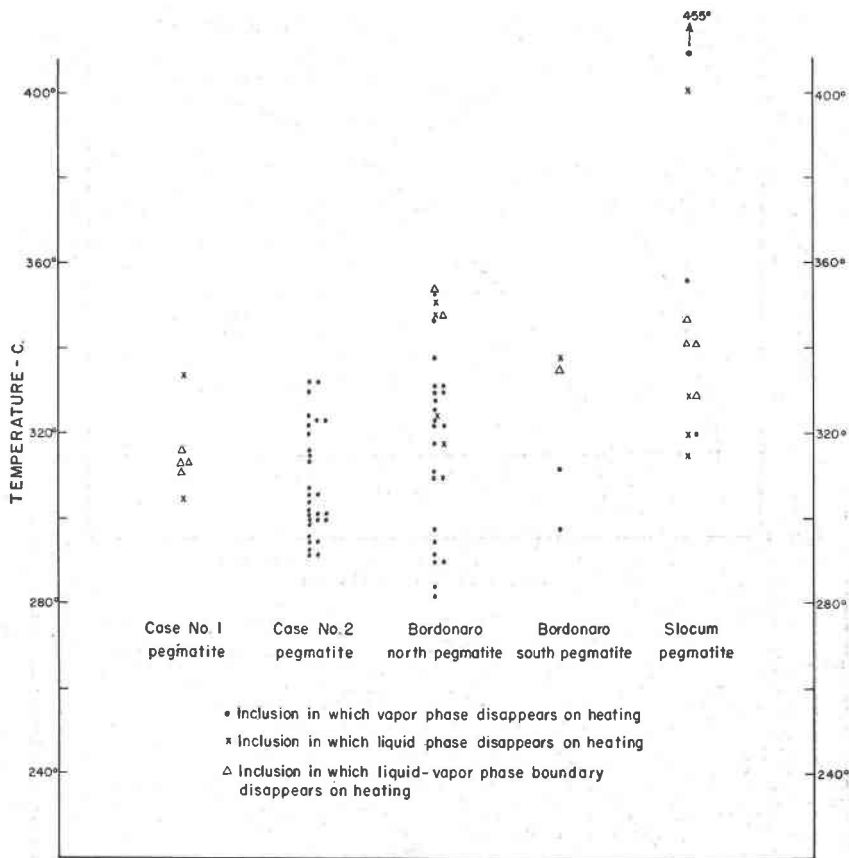


FIG. 9. Temperatures of disappearance of liquid phase, vapor phase, or liquid-vapor boundary in apparently primary three-phase inclusions in beryl from the Bordonaro north, Bordonaro south, Case No. 2, Slocum, and Case No. 1 pegmatites. Each symbol represents one inclusion.

pegmatite, and in two crystals each from the Slocum and Gotta-Walden pegmatites. Temperature data for these inclusions are also shown in Fig. 9, and may be summarized as follows:

<i>Pegmatite</i>	<i>Number of Inclusions</i>	<i>Temperatures of disappearance of the liquid phase</i>
Bordonaro north	3	324°-351° C.
Bordonaro south	1	338° C.
Case No. 1	2	305°-334° C.
Slocum	5	310°-401° C.
Gotta-Walden	15	310°-346° C.

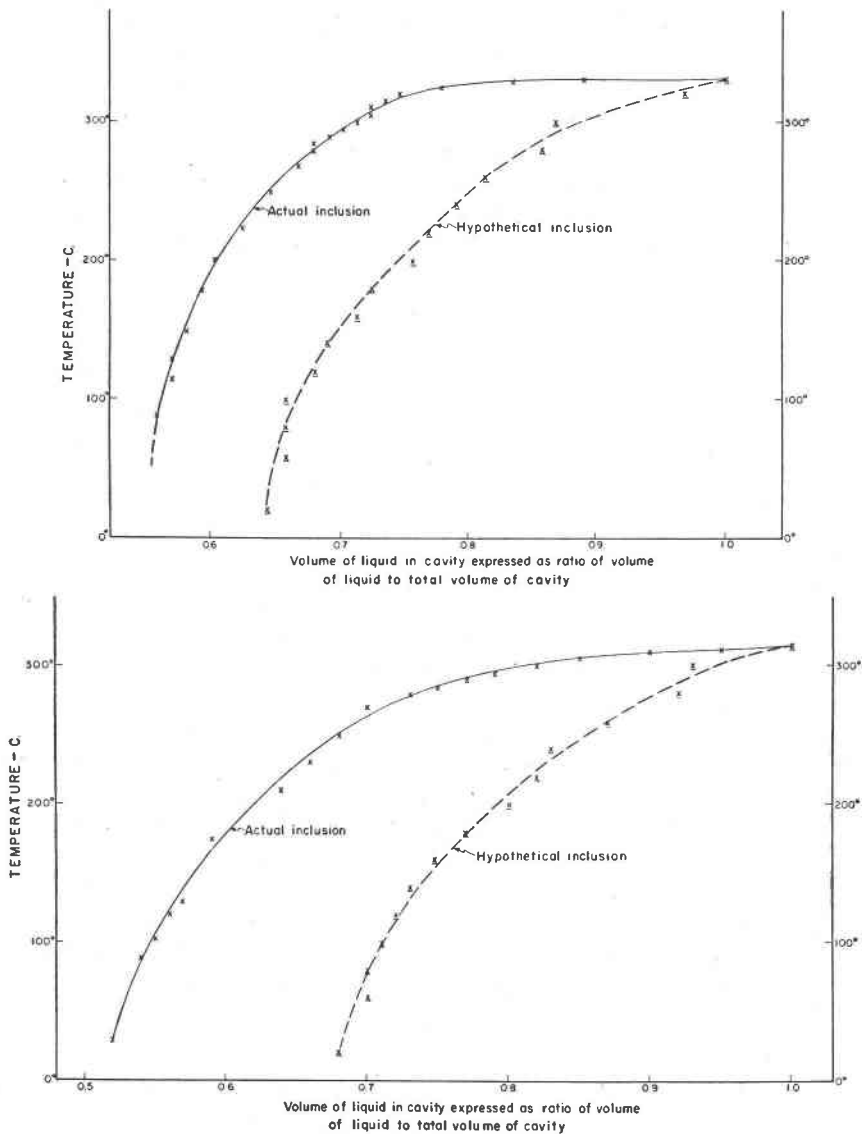


FIG. 10. Curves showing change in volume for actual inclusions of the three-phase type in beryl from the Case No. 2 pegmatite and for hypothetical inclusions composed of pure water. In each case, both the actual inclusion and the hypothetical inclusion are filled with liquid at the temperature corresponding to the upper end of the pair of curves.

In the second subgroup of inclusions, heating causes no change in liquid-vapor ratio, but the boundary between the two phases ultimately

fades and disappears as though the critical temperature of the liquid phase were reached. Ingerson has shown (1947, p. 378, Fig. 1) that this behavior is to be expected if an inclusion consisting of water and vapor in the proper ratio (about 3:7) at room temperature is heated to the critical temperature of water. In the inclusions under discussion the system involved is not so simple, but the principle involved is presumably the same.

Inclusions of the second subgroup have been found in two crystals of beryl from the Bordonaro north pegmatite, one crystal from the Bordonaro south pegmatite, and one from the Slocum pegmatite. Temperature data for these inclusions are shown in Fig. 9 and may be summarized as follows:

<i>Pegmatite</i>	<i>Number of Inclusions</i>	<i>Temperatures of disappearance of the phase boundary</i>
Bordonaro north	3	348°-354° C.
Bordonaro south	1	335° C.
Slocum	4	329°-347° C.

In the third subgroup of three-phase inclusions, progressive heating beginning at room temperature first causes the presumed liquid carbon dioxide phase to disappear, then the vapor phase shrinks perceptibly, then the liquid-vapor boundary fades and disappears. One inclusion of this sort has been found in a crystal from the Bordonaro north pegmatite. Presumably further search would disclose others, hence a possible subgroup consisting of such inclusions must be recognized. In the lone example, the phase boundary disappears at 354° C. It seems probable that this type of inclusion is a variant of the second subgroup, having a slightly different density.

Few data for the disseminated rounded, elongate, or irregular inclusions of type (2) have been obtained. Temperatures of disappearance of the vapor phase in four two-phase inclusions of type (2) in pink beryl from the Gillette pegmatite range from 232° to 263° C. Ten similar inclusions in beryl from the Strickland-Cramer pegmatite gave values of 180° to 260° C. In a three-phase inclusion in beryl from the wall zone of the Bordonaro south pegmatite the liquid phase disappeared at 383° C. The data are too limited to justify conclusions, but it is of interest that the values are similar to those of comparable apparently primary inclusions.

Data for only 18 inclusions of Type (3) are at hand. Fourteen inclusions in beryl from the Case No. 1 pegmatite show disappearance of the liquid phase at 287° to 316° C. The vapor phase in one inclusion in beryl from the Bordonaro north pegmatite disappeared at 322° C.,

in another at 335° C. The vapor phase in one inclusion in beryl from the Slocum pegmatite disappeared at 310° C., in another at 336° C. The determinations were made early in the work, when the present procedure for determining the presence of a third phase in inclusions at room temperatures was not yet worked out. Subsequent work has shown that most secondary inclusions in beryl from these two pegmatites are three-phase inclusions.

Certain broad relationships between behavior groups of apparently primary inclusions in beryl and the occurrence of beryl within the pegmatites studied appear to be suggested by the data. The data for the Case No. 1, Case No. 2, Bordonaro, and Slocum pegmatites, are for inclusions in beryl occurring in the wall zones or intermediate zones of the pegmatites. Plagioclase, perthite, and quartz are the essential constituents of these zones, and the zones apparently represent the early and middle stages of crystallization of the pegmatites. Apparently the characteristic inclusions in beryl having this occurrence are three-phase inclusions containing abundant liquid carbon dioxide. The data for the Selden east and Strickland-Cramer pegmatites, on the other hand, are for beryl from the last-formed units of the pegmatites, a pod in the former case, replacement bodies in the latter case. Apparently the characteristic inclusions in beryl in these late units are free of detectible amounts of carbon dioxide.

The Gotta-Walden pegmatite may be an exception to this two-fold grouping, for three-phase inclusions occur in beryl from the apparent core. It is doubtful, however, whether the true core of this pegmatite has ever been seen, either in surface exposures or in drill cores. The mineral assemblage forming the apparent core is the same as that forming wall or intermediate zones in the Case, Bordonaro, and Slocum pegmatites.

Fluid inclusions in quartz: Although quartz proved unsatisfactory for determinations of temperature by the fluid inclusion method, intensive study of a large number of plates prepared from quartz from the Case No. 2 and Case No. 1 pegmatites has yielded a few data. Three types of inclusions have been found, as follows:

- (1) Disseminated negative crystals.
- (2) Disseminated irregular inclusions.
- (3) Irregular to rounded inclusions arranged along cross-cutting planes or curved surfaces, many of which intersect or branch.

Inclusions of the first group are rare; those of the second group are not abundant. Inclusions of the third type are exceedingly abundant in every plate studied. In most of the plates cut, they are the only type of inclusion present. In the remainder, they generally outnumber the

other types of inclusions. Inclusions of the first two groups may well be primary, especially the negative crystals; inclusions of the third group are obviously secondary.

Determinations have been made successfully for 17 inclusions of the negative crystal type in quartz from the Case No. 2 pegmatite. In 15 of the inclusions, one from the wall zone and the other 14 from quartz of the core, the vapor phase first shrinks slightly but perceptibly, then the phase boundary fades and disappears. Temperatures of disappearance range from 293–357° C. In the other two inclusions, in a plate of quartz from the core, the vapor phase shrinks and finally disappears, at 352° C. in one inclusion, at 395° C. in the other.

Temperatures of disappearance of the vapor phase in 29 disseminated irregular inclusions in quartz from the core of the Case No. 2 pegmatite range from 124° C. to 292° C., but values for 27 of the inclusions range from 193° C. to 292° C. In 6 other inclusions from quartz of the core, and in one in quartz from the wall zone, the liquid-vapor boundary fades and disappears, at temperatures ranging from 320° C. to 390° C.

Four secondary inclusions of type (3) in quartz from the Case No. 1 pegmatite gave temperatures of disappearance of the vapor phase ranging from 316°–341° C. All other secondary inclusions of this type studied leaked at temperatures at which the liquid phase fell considerably short of filling the cavities.

Most of the inclusions of each of the groups described above are now known to consist of three phases at room temperature, like the inclusions in the associated beryl. A very few appear to consist of two phases. The remainder are so small that the presence or absence of a third phase can not be established.

DISCUSSION OF RESULTS

Assumptions involved in the use of fluid inclusions:

The use of fluid inclusions rests on a series of assumptions (Ingerson, 1947, pp. 376–377; Bailey and Cameron, 1951, p. 642). These must be reviewed with each new investigation of fluid inclusions, so that their validity with respect to the inclusions studied can be appraised. Review is particularly necessary for pegmatite minerals, because the physico-chemical conditions under which pegmatites crystallize are uncertain. The assumptions ordinarily made are as follows:

- (1) The cavity occupied by the inclusion was completely filled with liquid at the time of crystallization.
- (2) Primary fluid inclusions can be distinguished from secondary inclusions.
- (3) The liquid is an aqueous solution containing no carbon dioxide or other gas in large concentration.

- (4) The pressure on the liquid at the time of inclusion was small, or its magnitude can be estimated.
- (5) There has been no significant change in the volume of the cavity itself due to pressure, solution, or precipitation.
- (6) There has been no addition of liquid to the cavity or loss of liquid from it.
- (7) Representative samples are used for temperature determinations.

For liquid inclusions in pegmatite minerals, this is a formidable list of assumptions, quite apart from the results of the present investigation. Even the seventh cannot be accepted as justified for the beryl studied. The sections of crystals cut give a generous sampling of beryl in the pegmatites involved, except for the Gillette, Case No. 1, and Slocum pegmatites. Beryl studied from the Strickland-Cramer pegmatite is satisfactorily representative of beryl in the replacement bodies, though not of beryl in other units. However, many of the mineral sections cut contain no usable inclusions, and additional sections must be made before we can be sure that representative sampling has been attained.

The other assumptions are difficult matters. The first assumption can ordinarily be made with considerable assurance for inclusions in minerals that are known to have formed from solutions at low temperatures. This is not so for inclusions in pegmatite minerals. The physico-chemical nature of the fluids from which pegmatite minerals crystallize is decidedly uncertain, and there is no *a priori* assurance that the cavities in beryl were filled with liquid at the time the crystals formed. Depending upon one's ideas of the physical chemistry of pegmatitic fluids, the mother fluids of pegmatites may be regarded as consisting of a liquid phase below its critical temperature, a fluid phase above its critical temperature, a combination of a liquid phase plus a vapor phase (Bowen, 1933, pp. 115-118) or even (Smith, 1948*b*; Neumann, 1948) two immiscible liquid phases. Furthermore, it is conceivable that the number and kinds of phases (that is, whether vapor or liquid) present in the fluid may not be the same in the early stages of crystallization of pegmatites as in the later stages. The phases present will depend on relative concentrations of the various constituents, the hyperfusibles in particular, on the pressure within the system, and on the temperature.

How shall one distinguish among fluid inclusions produced under these various sorts of conditions? Let us assume that the pegmatite fluid is a liquid and is at a temperature below its critical temperature. Inclusions formed in crystals growing in such a liquid should fill their cavities at the instant of entrapment; the liquid phase should shrink and a vapor phase should appear and grow as the crystal cools to room temperature. If now the crystal is heated, the liquid phase should expand and ulti-

mately fill the cavity. This would be, then, an inclusion of the type normally envisioned in connection with use of the Sorby method. Among the inclusions here described, those in beryl from the Selden east, Strickland-Cramer, and Gillette pegmatites would, *so far as their behavior indicates*, appear to qualify as examples.

Let us suppose, on the other hand, that pegmatites form from fluids above their critical temperatures. The fluid entrapped will again consist of a single phase that will completely fill the cavity. Its behavior on cooling, however, will depend on its original density, which is a function of pressure. If its density exceeds the critical density, then as the crystal cools below the critical temperature, the inclusion will pass directly into a liquid phase that will completely fill the cavity. No second phase will appear until the cooling liquid shrinks so that it can no longer fill the cavity. At this point, a vapor phase will appear. When beryl containing such an inclusion is reheated, its behavior will be indistinguishable from that of an inclusion formed from a liquid below its critical temperature. Thus, using the data given by Kennedy (1950, p. 540, Fig. 3) both an inclusion formed of water at a temperature of 360° C. and at a pressure of about 440 bars and one formed at 555° C. and a pressure of 2120 bars would show disappearance of the vapor phase at 325° C. If we assume that the inclusions of the Selden east pegmatite are 5-molar aqueous solutions, then from the curve given by Ingerson (1947) for solutions of KCl and NaCl in water, the critical temperature of the inclusions would probably be greater than 400° C. In the absence of corroborative data, we cannot beg the question by assuming that pegmatite liquids do or do not attain such temperatures. The one conclusion possible is that regardless of whether inclusions of the type found in the Selden east pegmatite were entrapped at temperatures above or below their critical temperatures, the temperatures of disappearance of the vapor phase recorded for these inclusions are less than the true temperatures of crystal formation, provided the other assumptions underlying the Sorby method hold for these inclusions. If properly corrected for pressure, however, the temperatures of disappearance should give the temperatures of formation of the inclusions.

If the fluid trapped in an inclusion is above its critical temperature, but has a density less than the critical density, then three cases are possible. If the density is only slightly less than the critical density, then as the inclusion is cooled below the critical temperature, a liquid phase will appear. The liquid will not fill the cavity, however, hence a vapor phase will be present. As the crystal is cooled to room temperature, the liquid phase will shrink. This is the first case. The amount of shrink-

age will decrease with decreasing density of the inclusion at the time of entrapment, until a certain value of the density is reached for which the shrinkage of the liquid phase due to cooling is compensated by condensation of the vapor phase. This is the second case. In both cases, the inclusion at room temperature will consist of a liquid phase and a vapor phase. In the first case, the liquid phase will expand as the inclusion is heated, but before it expands sufficiently to fill the cavity, the liquid-vapor boundary will fade and disappear. In the second case, the liquid phase will neither shrink nor expand, but when the critical temperature of the liquid is reached, the liquid-vapor boundary will fade and disappear. In both cases, the temperatures of boundary disappearance will be less than the temperatures of crystal formation. The third case occurs when the density of the liquid at the time of entrapment falls sufficiently below that of the second case. In this third case, a liquid phase will appear and will expand as the inclusion is cooled to room temperature, and conversely, heating will cause the liquid phase to shrink and disappear at a temperature below the temperature of crystal formation.

If a fluid inclusion shows disappearance of the liquid phase or of the vapor phase, and the pressure is known, then appropriate correction for the pressure will give the temperature of formation of the inclusion, provided the inclusion satisfies the other assumptions of the Sorby method.

Finally, let us suppose that a beryl crystal happens to be forming at a time when minute bubbles of vapor are rising through a liquid phase. In such an environment, two kinds of inclusions may conceivably be trapped within a crystal, one type consisting of liquid only, the other consisting of liquid plus a bubble of vapor. Each will consist of a liquid phase and a vapor phase at room temperature. The first type, when heated, will show shrinkage and final disappearance of the vapor phase. If the inclusion satisfies the other assumptions of the Sorby method, the temperature of filling, corrected properly for pressure, will be the correct temperature of formation of the inclusion.

The behavior of the second type will depend on the ratio of liquid to vapor at the time of entrapment. For a certain critical ratio, the result will be an inclusion in which the liquid-vapor ratio is unchanged with cooling. When such an inclusion is heated, the phase boundary will not shift, but will fade and disappear when the critical temperature of the liquid is reached. The temperature of disappearance gives the critical temperature of the fluid composing the inclusion but gives no information as to the temperature at which the inclusion formed. If the liquid-

vapor ratio at the time of entrapment exceeds the critical ratio, the liquid will expand until it fills the cavity, but the temperature of filling, corrected for pressure, will be higher than the temperature of formation of the inclusion. If the liquid-vapor ratio is lower than the critical ratio, the vapor phase will expand as the inclusion is heated and will ultimately fill the cavity. The temperature of filling, corrected for pressure, will be either lower or higher than the true temperature of formation of the crystal, depending on the exact ratio (see Ingerson, 1947, Fig. 1 and p. 386).

If inclusions in a crystal have formed under such conditions, it seems probable that primary inclusions of all three types described above will occur together. If crystal after crystal within a given unit of a pegmatite shows all three or even two of these types, a two-phase mother liquor is a definite possibility. However, the same feature can also result from differential leakage of inclusions or from differential addition of material to inclusions.

The conditions outlined above, the possible types of inclusions formed, their appearance at room temperature, and their behavior when heated are summarized in Table 2. In the final two columns is given the occurrence of possible examples among the apparently primary inclusions in beryl that we have studied. Inclusions that consist of two phases at room temperature and those that consist of three phases are both included because for purposes of the foregoing analysis the composition of the inclusions makes no difference, and the existence of a third phase at low temperatures is extraneous to the discussion.

Several significant points have been brought out by compilation of the table. One is that all the types of apparently primary inclusions in beryl (and in quartz also) can be accounted for in terms of the possible phase conditions defined in the table. A second is that differences in degrees of filling and behavior of inclusions within a single crystal can be explained in terms of Case IV of the table without need of variations in either temperature or pressure within the period of formation of the crystal, and without need of differential leakage or differential addition. Explanation in terms of Case I, Case II or Case III, however, requires significant variations in temperature or pressure, or both, during the period of crystal formation, or requires differential leakage from, or addition to, the inclusions after their formation. A third conclusion is that nothing in the nature of a fluid inclusion at room temperature or in its behavior when heated tells us whether inclusions in which the liquid phase expands and fills the cavity belong under Case I, Case II, or Case IV; i.e., whether they have formed under supercritical or subcritical conditions

TABLE 2. KINDS AND BEHAVIORS OF POSSIBLE INCLUSIONS
IN PEGMATITE MINERALS

Nature of medium at time of crystal formation	Nature of possible fluid inclusions at time of entrapment			Nature of possible fluid inclusions at room temperature	
	Liquid	Gas	Liquid plus vapor	Liquid plus vapor	Liquid (1) plus liquid (2) plus vapor
I. Fluid, below critical temperature; no vapor phase present	x			x	
	x				x
II. Fluid, above critical temperature; density greater than critical density		x		x	
		x			x
III. Fluid, above critical temperature; density less than critical density	(1) Density near critical density			(a)	x
				(b)	x
	(2) Density moderately lower than critical density			(a)	x
				(b)	x
	(3) Density much below critical density			(a)	x
				(b)	x
IV. Liquid plus vapor	(1) Liquid only (2) Liquid-vapor ratio markedly higher than critical ratio	x	See under Case I	(a)	x
				x	
				(b)	x
	(3) Liquid-vapor ratio equal to critical ratio			(a)	x
				(b)	x
	(4) Liquid-vapor ratio less than critical ratio			(a)	x
(b)				x	

TABLE 2. KINDS AND BEHAVIORS OF POSSIBLE INCLUSIONS
IN PEGMATITE MINERALS (continued)

Behavior when heated					Occurrence of possible examples among inclusions studied	
Vapor phase disappears	Liquid (2) disappears then vapor phase disappears	Liquid phase(s) disap- pear(s)	Liquid (2) disappears (*) Liquid (1) expands some- what, then liquid-vapor boundary fades and disappears	Liquid (2) disappears (*) Liquid (1) vapor boundary fades and disappears	Pegmatite	Mineral containing the inclu- sions (†)
x					Selden east Strickland-Cramer Gillette Case #2	Beryl Beryl Beryl Quartz <i>D</i>
	x				Bordonaro north Bordonaro south Case #2 Slocum Gotta-Walden	Beryl Beryl Beryl Beryl Beryl <i>A</i>
x					Selden east Strickland-Cramer Gillette Case #2	Beryl Beryl Beryl Quartz <i>D</i>
	x				Bordonaro north Bordonaro south Case #2 Slocum Gotta-Walden	Beryl Beryl Beryl Beryl Beryl <i>A</i>
			x		None	
			x		Bordonaro north	Beryl
				x	None	
				x	Bordonaro north Bordonaro south Case #1 Slocum	Beryl Beryl Beryl Beryl <i>B</i>
		x			None	
		x			Bordonaro north Bordonaro south Gotta-Walden Case #1 Slocum	Beryl Beryl Beryl Beryl Beryl <i>C</i>
x					Selden east Strickland-Cramer Gillette Case #2	Beryl Beryl Beryl Quartz <i>D</i>
	x				Bordonaro north Bordonaro south Case #2 Slocum Gotta-Walden	Beryl Beryl Beryl Beryl Beryl <i>A</i>
				x	None	
				x	Bordonaro north Bordonaro south Case #1 Slocum	Beryl Beryl Beryl Beryl <i>B</i>
		x			None	
		x			Bordonaro north Bordonaro south Gotta-Walden Case #1 Slocum	Beryl Beryl Beryl Beryl Beryl <i>C</i>

(*) By these two columns, both inclusions consisting of two phases at room temperature and inclusions consisting of three phases at room temperature are covered. Heating of the three-phase inclusions first causes disappearance of one liquid phase, after which the inclusions behave as indicated by the headings of the columns.

(†) Each letter designates a particular group of inclusions. Thus the inclusions of group A cited as examples of Case I are the same as those of group A cited as examples of Cases II and IV (1) a.

from single-phase or two-phase systems. This fact is implied in part in discussions by Ingerson (1947), Scott (1948), Smith (1949), and Kennedy (1950) but has not been specifically stated.

A final conclusion is that even if one accepts the definitions of "hydrothermal" and "pneumatolytic" fluids proposed by Smith and his co-workers (1950, p. 587), the statement (p. 584) that

"If the liquid phase fills the inclusions before the vapor phase during heating, the transporting agent was a hydrothermal solution. Conversely, if the vapor phase fills the inclusion before the liquid phase during heating the transporting agent was a pneumatolytic mixture."

holds only if it can be shown that the system does not fall under Case IV of the table, and that there has been no leakage or addition.

Table 2 indicates that even if inclusions can be shown to satisfy other assumptions underlying the Sorby method, it is by no means easy to interpret temperature data with reference to the first assumption. When the validity of the other assumptions is appraised, difficulties multiply at a disconcerting rate.

The second assumption, that primary inclusions can be distinguished from secondary inclusions, is far less easy to satisfy than a perusal of the literature would indicate. Criteria for distinguishing primary from secondary inclusions have been discussed by Laemmlein (1929). He gives the following as characteristics of primary fluid inclusions:

- (1) They are always associated with real crystal surfaces.
- (2) They may be arranged parallel to growth planes of a crystal, or along a surface that records the direction of growth of an edge of a crystal; or along a row or line that traces the growth direction of the apex of a crystal.
- (3) They may consequently show a zonal arrangement corresponding to the growth zones of a crystal.
- (4) When arranged along planes, these planes do not intersect.

By contrast, he found experimentally that whereas some secondary inclusions formed by healing of fractures are negative crystals, others are rounded, oval or irregular. He pointed out that fluid inclusions formed by partial healing of fractures occurred along planes that cut across growth zones and other primary features of crystals and may branch or intersect. Laemmlein's discussion does not take fully into account the formation of primary inclusions along lineages.

In his discussion of secondary inclusions, Laemmlein appears to have considered only their development by the partial healing of cracks in crystals. Little consideration has been given to the possibility that fluid inclusions, negative crystals, or rounded or irregular inclusions may be of secondary origin even if they are disseminated through a crystal and appear unrelated to cracks or other features that might offer channelways

for later solutions or might influence selective solution. Ingerson (1947, p. 377) concluded that one is probably warranted in assuming that inclusions in crystals that line vugs and other open spaces in veins are primary. He regarded inclusions along planes that cross grain boundaries without offset or change in orientation as almost certainly secondary, and stated that if planes of inclusions have a more or less constant orientation or orientations over an entire thin section, the chances are good that the inclusions are secondary, even those in planes not observed to cross grain boundaries. He further stated that where planes of inclusions are confined to individual mineral grains or where arrangement in planes is not evident, the chances are better that the inclusions are primary. He also pointed out that though control of orientation by the crystal lattice of grains can be evidence that inclusions are primary, secondary inclusions may also be thus controlled. Disseminated inclusions, related or unrelated to crystallographic elements of the host crystal, are not discussed in his article, nor is the question of irregular inclusions versus negative crystals discussed. He figures, however, two irregular inclusions in quartz that are presumably of the disseminated type. Twenhofel (1947), based his determinations of temperature on inclusions that apparently are arranged along growth zones of a crystal. There seems little doubt that the inclusions he studied were primary. Those who have developed and employed the decrepitation method (Scott, 1948; Peach, 1949; Smith, 1948*a*; Peach, 1951) have recognized the importance of distinguishing primary from secondary inclusions, but have discussed the means of distinguishing the two only briefly; indeed, the difficulty of interpreting the results of decrepitation in terms of primary and secondary inclusions appears to the writers to be a basic weakness of the decrepitation technique, and our results for quartz from the core of the Case No. 2 pegmatite confirm this view in one instance. Peach (1951, p. 34) states that secondary inclusions in pegmatite minerals were recognized by the relatively small volume of the bubbles and by their tendency to occur in planes within the mineral. Primary inclusions were recognized by their random distribution and lower degree of filling. Are these acceptable criteria? The writers would be reluctant to accept the highly irregular inclusion in quartz figured by Peach (1951, Plate 1, *A*) as primary without corroborative evidence. We have found secondary inclusions in quartz and other minerals that are remarkably similar. Buerger concluded from a study of lineages in crystals (1932*a*, 1932*b*), however, that primary cavities of various shapes, some highly irregular, are common along lineage structures in crystals.

A perusal of the literature suggests that the "apparently primary" inclusions in beryl and quartz discussed in the present paper would be

accepted as primary without question by at least the majority of investigators, and that the disseminated irregular inclusions would likewise be accepted as primary. Furthermore, we find no record of experiments showing that such inclusions can develop by secondary processes. Nevertheless, we feel that concrete proof of primary origin of the inclusions is necessary before conclusions based on them can be fully accepted. The student of ore minerals knows all too well that disseminated solid inclusions can be developed in ore minerals by replacement, and selective sericitization and other alterations in zoned feldspars are likewise familiar to the petrographer. Perhaps liquid inclusions can develop in a similar manner.

The best evidence for primary origin of fluid inclusions appears to be as follows:

- (1) Inclusions distributed or oriented with reference to growth zones or other primary features of crystals and showing no relation to cracks or other secondary features.
- (2) Marked contrast in occurrence and in liquid-vapor ratios to clearly secondary inclusions in the same crystals.
- (3) A systematic pattern of temperature variation from part to part of a crystal, or from mineral to mineral within a deposit, that is consistent with paragenetic sequence and with the geologic characteristics of a deposit.
- (4) Clear cut evidence, in the form of cross-cutting relationships, that the inclusions were present before the formation of later pegmatite minerals; e.g., transection of inclusions by the walls of veinlets formed by later minerals.
- (5) Agreement between temperatures of formation, as determined from the inclusions, and other geologic evidence.

If a group of fluid inclusions satisfies *both* (1) and (2) above, the probability that they are of primary origin is high. If a certain type of inclusion is present in crystal after crystal from a given deposit, and shows a consistent contrast in characteristics to inclusions related to secondary features, the chances that they are secondary are slight (Bailey and Cameron, 1951, p. 643). The case for primary origin becomes even stronger if the results of investigation satisfy conditions (3) and (5). It must be recognized, however, that only condition (4) constitutes unequivocal proof of primary origin. So far as the writers know, no case of this kind has been reported in the literature.

From the foregoing considerations, it seems necessary to conclude that primary and secondary inclusions are difficult to distinguish even in transparent minerals, in which the characteristics and distribution

of inclusions can be studied. Such study is impossible for opaque minerals, and much though we admire the ingenuity of the decrepitation technique, this is a basic weakness of such fundamental importance that it makes all temperature data based on decrepitation of opaque minerals suspect of serious error.

The third and fourth assumptions can be considered together. Temperature data obtained by study of liquid inclusions require correction for the pressure at time of crystal formation. As Kennedy (1950, p. 543) has pointed out and others have recognized, this correction is based on the assumption that the coefficient of thermal expansion and the compressibility of the liquid in a fluid inclusion are essentially the same as those of pure water. This assumption is commonly forced on investigators because the compositions of fluid inclusions are little known and because temperature-pressure-volume data for solutions of various compositions at high temperatures and pressures are not available. It is generally assumed that fluid inclusions are dilute aqueous solutions, that the compressibilities of these solutions are not far different from those of pure water, and that the pressure corrections read from various charts showing P - V - T relationships for pure water (Ingerson, 1947; Scott, 1948; Kennedy, 1950) are of the right order of magnitude. This piling of assumption on assumption is certainly one of the weaknesses of the Sorby method. The errors thus introduced cannot be evaluated accurately at present. For deposits formed at shallow depths, such as the lead-zinc deposits of the Upper Mississippi Valley (Bailey and Cameron, 1951), pressure corrections required must be small and the errors introduced not serious. Both Ingerson (1947, p. 386) and Kennedy (1950, pp. 542-543) point out that for the deeper-seated deposits, however, errors of 50 per cent or more may be involved. If a gas of low critical temperature, such as CO_2 , is present, no pressure correction can be calculated, because data for systems consisting of H_2O plus CO_2 , H_2S , and other gases at high temperatures are lacking.

Even if an inclusion is known to consist solely of water, the usual pressure correction based on the P - V - T tables for water can be applied only if we know which of the four cases of Table 2 is represented. This may or may not be possible. Inclusions in minerals from deposits regarded on geologic evidence as formed at low temperatures generally show filling of the cavity by the liquid phase when the inclusions are heated, and it is probably safe to assume that these fall under Case I. The deeper-seated the conditions under which a deposit has formed, however, the greater is the uncertainty. For pegmatites, as we have seen, the uncertainty is very great.

From the standpoint of pressure correction, it makes no difference

whether Case I or Case II is involved. The problem is to recognize whether either is involved. If all inclusions in a crystal or growth zone of a crystal show disappearance of the vapor phase on heating and show the same liquid-vapor ratio, the presumption is strong that they fall under Case I or Case II, although it is still possible that the inclusions have undergone uniform change in content (see below).

Similarly, if all the inclusions of a crystal or a growth zone of a crystal show *one* of the types of behavior described under Case III of the table, the presumption is strong that the inclusions fall under this case. Pressure corrections for this case are not given by ordinary curves, but can be calculated from the liquid-vapor ratios at room temperature if the pressure at time of formation is known.

Inclusions falling under Case IV, as discussed previously, should show a variety of behavior. A pressure correction can be applied only to those inclusions that consisted wholly of liquid at the time of entrapment. These would be the inclusions showing disappearance of the vapor phase at the lowest temperature.

There is still the problem of estimating pressure. As pointed out by Ingerson (1947, p. 386) and Kennedy (1950, p. 542), we rarely can determine whether this should be taken as the hydrostatic pressure, the pressure due to the weight of overlying rocks, or local pressures. Pressure at time of crystal formation is generally taken as equivalent to the weight of overlying rocks (e.g., Ingerson, 1947; Peach, 1951), but it is recognized that this is a statistical view of the problem. Smith and his associates (Smith, 1948*a*; Peach, 1951) rely on the pyrite geothermometer in calculating pressure corrections to be applied to temperature values obtained by decrepitation. The method is subject to whatever errors may be inherent in the technique, plus possible serious errors arising from the fact that neither the nature nor the origin of the inclusions presumed to be responsible for decrepitation in pyrite can be determined.

For the inclusions in beryl and quartz discussed in the present paper, no clear-cut estimate of pressure at time of formation can be made. If the pressure be taken as due to the weight of the overlying rock, then a pressure corresponding to a depth of 15,000 feet is probably to be regarded as a minimum. We must recognize, however, that the actual pressure may be unrelated to thickness of rock cover.

The fifth assumption underlying the Sorby method concerns possible changes in the volume of the cavity itself due to pressure, to solution of the walls, or to precipitation of solute upon the walls. This subject has been discussed by Ingerson (1946, pp. 376-377) and by Bailey and Cameron (1951, pp. 644-645). It seems probable that errors introduced in this way are minor.

The final assumption to be discussed is that involving addition or loss of fluid or vapor since the inclusions were formed. This is a question that has been given emphasis by Kennedy's discussion (1950), which is backed by experimental evidence that the fluid content of inclusions in fluorite can be increased by immersion in water under high pressure. Increase in fluid content lowers the temperature of disappearance of the vapor phase, whereas decrease in fluid content increases the temperature. It is not necessary, moreover, that there be change in quantity of fluid but only a change in composition, for the compressibility and coefficient of thermal expansion of the fluid will thereby be influenced even if the quantity of fluid remains unchanged.

It is obvious that no definite answer can be given to this question. Kennedy reasons that in veins the fall in temperature of the enclosing rocks is probably more rapid than the fall in pressure, and that the tendency will be for migration of material into fluid inclusions, so that temperatures of disappearance of the vapor phase will be lowered. Whether this is the case for pegmatites is largely a matter of opinion; we have no direct knowledge of relative rates of decrease of temperature and pressure after the formation of pegmatites. Furthermore, we know little of the pressure gradients required to induce migration of material or of the rates of migration. The pressure gradient of 1500 bars employed in the experiments cited by Kennedy was extreme. Kennedy's point, however, is one that merits consideration.

Smith and his co-workers (Smith *et al.*, 1950, p. 584) have pointed out that if adjustment of the contents of fluid inclusions to changes in pressure environment is an effective process, then it is difficult to explain why quartz from gold deposits of the Canadian shield contains two sets of inclusions exhibiting different degrees of filling. It might therefore be argued that because primary and secondary inclusions in quartz from the Case No. 2 pegmatite are similar in degrees of filling, the contents of the primary inclusions have been changed. Field study of the pegmatite suggests that the secondary inclusions could readily have formed at a later stage in the development of the pegmatite, however, and it is entirely possible that they developed within the same temperature-pressure range.

Two features of the inclusions described in the present study, however, require mention. For the carbon dioxide-bearing inclusions the pressure gradient is at present the reverse of that envisioned by Kennedy, for the pressure within the inclusions at room temperature must be close to the critical pressure (73 atm.) of carbon dioxide.

The second feature is the occurrence of apparently primary inclusions of different degrees of filling within a single crystal, in places within a cubic millimeter. This is difficult to explain if material has migrated

in response to an external pressure gradient, unless there are complicating factors.

Further than the above, the effects of migration of material are difficult to appraise. It may well be that the spread of temperature values shown by inclusions in certain crystals or by various crystals from a single pegmatite is due partly or wholly to this cause.

A final source of error is one pointed out by Dreyer, Garrels, and Howland (1949). Owing to exothermic reactions and poor conduction of heat away from the surface of a growing crystal, the temperature of a liquid inclusion may exceed that of the surrounding liquid. This is a difficult problem for the student of pegmatites, because neither the reactions involved in crystallization nor the rates of heat conduction are known. However, crystallization from a solution is itself an exothermic reaction, hence heating of the liquid immediately adjacent to the face of the growing crystals is a definite possibility.

INTERPRETATION OF THE DATA

General Statement: It is evident that there are numerous possible sources of error in interpreting temperature data for pegmatite minerals. If the oriented tubular inclusions and negative crystals are not primary, the temperature values obtained are not related to temperatures of crystal formation, but to temperature conditions during some later period in the history of the pegmatites. The cavities in some of the inclusions may not have been filled with liquid at the time they formed. No firm estimate of pressure at time of formation can be made, and since the pressure-volume-temperature relationships for the three-phase inclusions are unknown, the pressure corrections could not be calculated even if the pressures were known. The possibility that the cavity contents have changed due to movement of material in or out of the cavities cannot be evaluated. Exothermic effects at the faces of the growing crystals are likewise possible.

With so many variables to consider, an unique interpretation of the data in terms of temperatures of crystal formation is impossible. The question is: Can we recognize limits of temperature within which crystallization must have taken place? The discussion that follows is addressed to this question. The "apparently primary" inclusions are the ones chiefly considered; it is assumed for purposes of the discussion that these inclusions either are truly primary or were formed at some stage of the development of the host pegmatite body, hence are significant with respect to conditions of pegmatite formation.

Two-phase inclusions in beryl: Suppose we consider first the apparently primary inclusions in beryl that consist of two phases at room tempera-

ture. How can the broad range of temperatures of disappearance of the vapor phase shown by inclusions from the Selden east pegmatite be explained? Values for 86 inclusions are given in Fig. 6. All show disappearance of the vapor phase, and hundreds of others with comparable degrees of filling have been observed in the crystal sections studied. The 36 inclusions of this type in beryl from the Strickland-Cramer pegmatite shown in Fig. 6 show the same behavior. The inclusions therefore cannot fall under Case III of Table 2. It seems unlikely, moreover, that the inclusions fall under Case IV of Table 2, for if this were so, a few, at least, would show lower degrees of filling, and either disappearance of the vapor phase or critical phenomena. For the same reason, differential leakage seems unlikely, for Case IV actually describes the behaviors expectable if inclusions formed under conditions of Case I or Case II have leaked differentially. If, however, the curves for these inclusions are similar to those given for water and a 10 percent KCl solution by Ingerson (1947, Fig. 1), inclusions showing critical phenomena or disappearance of the vapor phase would not be developed unless leakage reached an advanced stage. It is therefore possible either that the inclusions fall under Case I or Case II and that the range of values is due to leakage, or that they fall under Case IV. In either case, the true temperature of formation, uncorrected for pressure, should be nearest the lowest value given by any of the inclusions; namely, 187° C. for beryl from the Selden east pegmatite and 194° C. for beryl from the Strickland-Cramer pegmatite. The same would hold if we regard the variation in temperature as due to exothermic effects at the faces of the growing crystals.

On the other hand, let us consider the possibility that material may be added to the inclusions after their formation, as Kennedy suggested. If the inclusions were formed under conditions of either Case I or Case II, addition of fluid would account for the variations in degree of filling, and also for the fact that none of the inclusions has so low a degree of filling that it shows either critical phenomena or disappearance of the vapor phase. If this is the cause, however, then the true values of temperatures of formation of beryl, uncorrected for pressure, should be nearest the highest temperatures of disappearance; namely, 268° C. for the Strickland-Cramer pegmatite and 385° C. for the Selden pegmatite. From their geologic characteristics, such a marked contrast in temperatures of formation would hardly be expected for the two pegmatites, but the reader may well object that additional temperature studies for the Strickland-Cramer pegmatite might narrow the gap. Kennedy's suggestion is a definite possibility.

Still a third approach is the statistical one advocated by Smith and

his co-workers (Smith *et al.*, 1950, p. 587). By this, the temperature of formation of beryl in the Selden east pegmatite, uncorrected for pressure, should be taken as somewhere between 210° and 290° C., the range in which most of the values lie. We might find support for this interpretation in the fact that where groups of inclusions in a crystal give closely similar values, these lie within the range above, and in the further fact that most of the values for beryl from the late-formed units of the Strickland-Cramer pegmatite and the Gillette pegmatite likewise lie in the same range. With so many possible sources of variation in temperatures of disappearance, can we accept a statistical norm? We question this, because the preceding analysis of the possible sources of variation in two-phase liquid inclusions suggests that if a group of inclusions shows a range of degrees of filling, the degree of filling appropriate to the temperature of formation is more likely to be represented by one or the other end of the range than by the median values.

This argument breaks down, however, when we consider that several processes may be jointly responsible for variations in degree of filling of a group of inclusions. One or more processes might increase the degree of filling of some of the inclusions, another process or processes might decrease the degree of filling of still other inclusions. In this case, the proper degree of filling (and temperature of vapor phase disappearance) would be between the two extremes found. But what criteria have we that will enable us to diagnose such cases?

The writers conclude that for the present, the temperature of formation of the beryl in the Selden east pegmatite, uncorrected for pressure, can only be taken as lying within the range 187° C. to 386° C. If the pegmatites formed under rock pressures corresponding to depths of 3 miles, and the compressibilities and thermal characteristics of the inclusions approach those of pure water, the corresponding temperatures of formation would be roughly 272° C. and 600° C. Actually, however, there is no means of determining the true pressure. So far as the data indicate, beryl formed in the Strickland-Cramer and Gillette pegmatites within the same range of temperature as the Selden east pegmatite.

Three-phase inclusions in beryl: If two-phase inclusions present such difficulties, what of the three-phase inclusions, which are even more complex? Let us consider the data for three-phase inclusions in beryl from the Bordonaro north pegmatite, as given in Fig. 11. Various inclusions in beryl from the pegmatite show three types of behavior when heated—(1) filling with a liquid phase, (2) filling with a vapor phase, or (3) disappearance of the phase boundary. Taken individually, these behaviors might result, for (1), under Case I, Case II, or Case IV; for (2) under Case III or Case IV; and for (3) under Case III or Case IV. Our problem, however, is not to account for any single behavior type,

but for the entire assemblage. In this connection, the frequency of occurrence of the three behavior types is of interest. The figure shows that of 28 inclusions for which precise data are available, 23 show filling with liquid, three show filling with vapor, and only two show fading of the phase boundary. Inasmuch as all three types are found in a single crystal, and the occurrence of the three types is evidently unrelated to stages of crystal growth, it seems unreasonable to account for

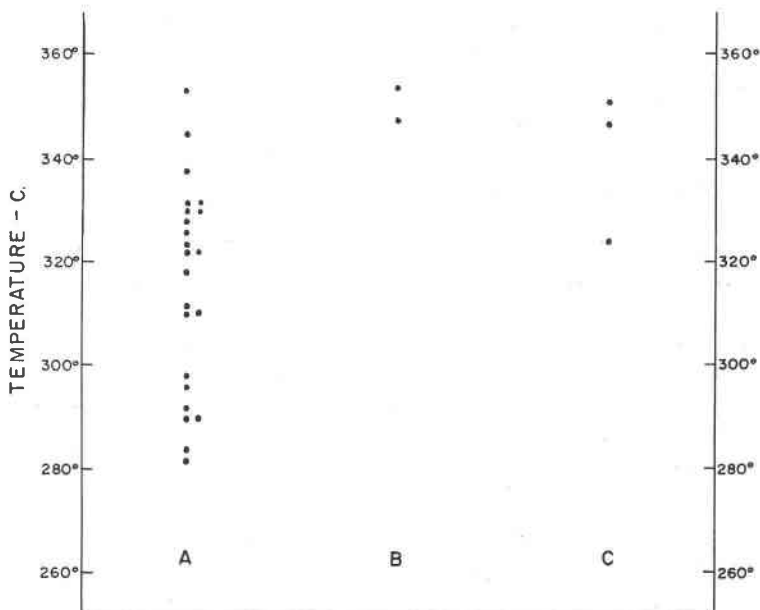


FIG. 11. Temperature data for inclusions in beryl. Bordonaro north pegmatite. Each symbol represents one inclusion. Heating causes disappearance of the vapor phase in inclusions of group *A*, disappearance of the liquid-vapor boundary in inclusions of group *B*, and disappearance of the vapor phase in inclusions of group *C*.

the three types on the basis of variations in over-all phase conditions in the mother liquid. Several possibilities remain, however. First, the inclusions may have formed under conditions of Case IV. The true temperature of crystal formation uncorrected for pressure, should then be closest to the lowest value found; namely, 282° C. Second, the variation may be due to differential leakage of inclusions originally formed under Case I or Case II; again the lowest value should be taken. The same would hold true if the variation is due to exothermic effects at the faces of growing crystals; it is necessary to assume, of course, that local heating was enough to produce a vapor phase.

The relationships shown in Fig. 11 are consistent with any of these

possibilities. Reference to the work given by Ingerson (1947, Fig. 1), together with rough measurements of degrees of filling in the inclusions in beryl from the Bordonaro pegmatite, indicates the following:

- (1) Inclusions showing disappearance of the liquid phase fall within a narrow range of degrees of filling, hence should be relatively few in number, whereas those showing disappearance of the vapor phase fall within a broader range, hence should be relatively numerous. Inclusions showing fading of the phase boundary must have degrees of filling falling within an extremely narrow range, and should be rare.
- (2) The temperatures of disappearance of the vapor phase, or of the liquid phase, should not exceed the temperature of disappearance of the phase boundary. This corresponds to Fig. 11, except that one inclusion showed disappearance of the phase boundary at a temperature 6° below the maximum. As has been noted, however, the precise point at which fading of a phase boundary is complete is extremely difficult to determine in such tiny inclusions, and temperatures recorded are considerably less accurate than those recorded for disappearance of liquid or vapor.

The final possibility to be considered is that the variation is due to addition of material to the inclusions. If this be so, the inclusions must have formed under the conditions of Case III [(3)*b*], otherwise we cannot account for the inclusions that show disappearance of the vapor phase or fading of the liquid-vapor boundary. The lowest value of the degree of filling should then be nearest the original value, and the temperature of disappearance of the vapor phase in this inclusion should be nearest the temperature of formation of the inclusion, uncorrected for pressure. For the Bordonaro north pegmatite (Fig. 11) this temperature would be 324° C. Again, as under Case IV, and as in the cases of leakage or exothermic heating, the temperature of disappearance of the vapor-liquid boundary in inclusions showing this behavior should be greater than the temperature of disappearance of the vapor phase in any of the inclusions of group *A*, Fig. 11, or the temperature of disappearance of the liquid phase in inclusions of group *C*.

There is nothing in the characteristics of the inclusions in beryl from the Bordonaro pegmatite that would indicate which of the above is the true explanation. For possible clues, let us consider the data available for three-phase inclusions in beryl in other pegmatites. The data for the Bordonaro south pegmatite are too scant to have much significance, though the fact that results are in the same range as those for the Bordonaro north pegmatite is of interest. For the Case No. 2 pegmatite,

(Fig. 9), data for 28 inclusions are available; these inclusions were selected, on the basis of estimates of degree of filling, as representative of hundreds of similar apparently primary inclusions. Temperatures of disappearance range from 292° to 332° C.; this range has its limits within the broader range of values shown by the Bordonaro pegmatite. However, we are here faced with the same difficulties presented by the two-phase inclusions. If Case IV of Table 2 applies, or if leakage is the cause of variation, why is it that none of the inclusions shows disappearance of the liquid phase? Addition of material and exothermic effects remain as possible explanations, the one pointing to acceptance of the highest value as most closely approximating the proper temperature of disappearance, the other pointing to the lowest value. Data for the Gotta-Walden pegmatite present somewhat the same problem as those for the Bordonaro north pegmatite. The upper limit of temperature values is about the same, and all but three values are within the same range. These three are lower, the lowest 238°. Data for the Slocum and Case No. 1 pegmatites are inadequate, though of interest because the values are comparable in general to those obtained for three-phase inclusions in beryl from other pegmatites. Two inclusions in beryl from the Slocum pegmatite, however, give values of 400° C. and 455° C., respectively.

We are thus left to conclude, pending further investigation, that no choice among alternative explanations of the variations in degrees of filling is justified.

Certain comments appear warranted, however, by the available data. The Bordonaro north, Bordonaro south, Case No. 1 and Case No. 2 pegmatites are closely comparable in mineral composition and in structural characteristics, and the beryl studied comes from the same or closely allied mineral assemblages in each pegmatite. The Slocum pegmatite is similar, though the beryl is golden instead of green. The Gotta-Walden pegmatite is structurally different, but the beryl is from the same general mineral assemblage in which beryl occurs in the other five pegmatites. For all six pegmatites, the ranges of values are much the same, and in all six the apparently primary inclusions are of the three-phase type. This suggests (1) that the inclusions are indeed primary, (2) that the temperature data have significance in terms of temperatures of formation, and (3) that the key to the problem is a sound basis for choosing among the various possible causes of variation in the behavior of the inclusions.

Another point worthy of note is the range of critical temperatures indicated for the three-phase inclusions, as follows:

Bordonaro north pegmatite	348°–354° C.
Bordonaro south pegmatite	345° C.
Case #1 pegmatite	311°–316° C.
Slocum pegmatite	329°–347° C.

The ranges are narrow in a given pegmatite, and in all pegmatites the critical temperatures are below those of pure water.

Data for the system H_2O-CO_2 at high temperatures and pressures are not available, hence no basis for calculating pressure corrections for three-phase inclusions exists. However, Kennedy's statement (1950, p. 543)

"if the solution in the vacuoles contains dominantly dissolved gases, such as CO_2 and H_2O , or volatile fluorides with low critical temperatures, the pressure correction may be greater than for pure water."

is applicable.

Secondary inclusions in beryl: Further work on secondary inclusions in beryl is needed, but a few comments appear warranted at the present stage. The chief points of interest are (1) that results to date give temperature values comparable to those for the apparently primary inclusions, and (2) that if the apparently primary inclusions in a crystal consist of three phases, the secondary inclusions commonly show three phases likewise. The implication is that the secondary inclusions formed from solutions of the same general composition as the apparently primary inclusions and probably only a short time after the host crystals developed; i.e., within the period of pegmatite formation.

Inclusions in quartz: Data for inclusions in quartz from the Case No. 2 pegmatite are impossible to interpret at the present time. The type most likely to be primary, the disseminated negative crystals, gave data so erratic that analysis of the type applied to the inclusions in beryl yields no rational results. Fifteen inclusions showed disappearance of the liquid-vapor phase boundary at temperatures of $293^\circ C.$ to $357^\circ C.$ Of these, 14 were found in two specimens of quartz from the core of the pegmatite. One of these specimens contained an inclusion showing disappearance of the vapor phase at $395^\circ C.$, a temperature higher than the apparent critical temperature of any of the 14 associated inclusions. Variation in the composition of this group of inclusions is therefore suggested.

The contrast in behavior between the disseminated irregular inclusions, in which the vapor phase disappears at temperatures of $124^\circ C.$ to $292^\circ C.$, and the disseminated negative crystals is a noteworthy feature, but its meaning is uncertain.

The data for obviously secondary inclusions are too scant to be significant. The great abundance of these inclusions, which are so numerous that they give the specimens a milky, banded appearance, is worth emphasizing. Although they likewise commonly consist of three phases at room temperature, it may be that during their formation the other types of inclusions were irregularly modified.

One of the two specimens of core quartz cited above was Cameron's specimen No. 46-C-3, which was studied earlier by Ingerson (1947, Table 2, No. 5) and Scott (1948, p. 650). Ingerson obtained 155° C. for the probable best value of the temperature of disappearance of the vapor phase in inclusions in this specimen, and Scott obtained a value of 165° C. by decrepitation. Dr. Ingerson kindly returned the specimen for our use, together with his notes. His measurements appear to have been made on disseminated irregular inclusions, most of which are now known to consist of three phases at room temperature. The data here given show that we have been unable to duplicate his results, for although one of our 29 inclusions of this type gave a value of 124° C. and another 160° C., all others gave considerably higher temperatures. In addition, the disseminated negative crystals, which seem more likely to be primary than the others, give values that are much higher than 155° C. Finally, in all specimens of beryl examined by us, including a crystal from the core, the apparently primary inclusions are of the three-phase type and show disappearance of the vapor phase at 292° C. or above, more than 100 degrees above the temperature of 175° C. found by Ingerson for an inclusion in a small crystal of beryl present in specimen 46-C-3.

The value of 165° C. obtained by Scott probably represents bursting of secondary inclusions. These are so exceedingly predominant that their bursting would be expected to mask the bursting of the other types of inclusions. The point of steepening of the decrepitation curve for this specimen (Scott, 1948, Fig. 4c) corresponds quite well with the temperature at which we found fracturing and leakage of inclusions to begin when the plates of the quartz were heated on the microscope stage. There is some evidence, however, that the inherent strength of quartz is the controlling factor, rather than the degrees of filling of inclusions.

Composition of the liquid inclusions and the origin of pegmatites: It has generally been assumed that primary liquid inclusions in minerals are accurate samples of the mother liquors from which the host minerals were deposited, and some attention has been given to analysis of inclusions as a guide to the composition of mineral-depositing fluids (e.g., Newhouse, 1932; Buerger, 1932b). More recently, Roedder (1951) has discussed the results of analysis of inclusions in pegmatitic quartz. He finds that the inclusions are solutions having concentrations of 10 per cent or more. The ratio of K to Na was found to be about 1 to 1, and the ratio of Cl⁻ion to SO₄⁼ion (sulfide ion would appear here in the analysis) was found to range from 0.5 to 50. The findings suggest moderately concentrated aqueous solutions.

As reported previously (Cameron, Rowe, and Weis, 1951), the fluid inclusions studied, both those in beryl and those in quartz, are highly

mobile at room temperatures, and this together with the heating curves for the solutions, the results of freezing experiments, the disappearance of one liquid phase of the three-phase inclusions, and the critical phenomena, suggest strongly that inclusions in beryl in some of the pegmatites are aqueous solutions and that those in beryl and quartz in other pegmatites are similar but richer in carbon dioxide. The inclusions do not have the properties that would be expected of solutions rich in silicates.

The nature of these inclusions has a definite bearing on the problem of the origin of pegmatites. Some of the beryl and quartz studied came from the wall zones and outer intermediate zones of the pegmatites. Let us suppose, for purposes of argument, that the inclusions are truly primary and that they are true samples of the mother fluids from which the pegmatites crystallized. Then if pegmatites crystallize from magma in closed or restricted systems (Cameron, Jahns, McNair, and Page, 1949, pp. 98-105), the primary inclusions in minerals of outer zones should be rich in constituents of the inner zones. Furthermore, as compounds of these constituents should be relatively insoluble at room temperature, the inclusions should consist of crystals of silicates as well as fluids. If such inclusions are lacking, then the pegmatites must have formed in open systems from fluids of hydrothermal or pneumatolytic character.

This matter merits thorough investigation. It is not a simple one. First, the assumption that the inclusions are representative samples of the mother liquors needs examining. The validity of the assumption for large primary inclusions in minerals is hardly debatable, provided post-depositional modification can be excluded. However, for inclusions measurable in microns, as are all those described in the present paper, this assumption needs checking by laboratory experiments in growing inclusions in polycomponental solutions. Concentration gradients are known to exist in liquids in which crystals are growing (Bunn, 1949). The microns-thick layer of fluid immediately adjacent to the growing face of a crystal may or may not have the same composition as the surrounding liquid.

A second possibility has been pointed out by Professor R. M. Garrels, of Northwestern University (oral communication). Neumann (1948) and Smith (1948*b*) have suggested that immiscibility occurs in pegmatitic liquids, resulting in a predominant silicate-rich liquid and a subordinate water-rich liquid. It is possible that the fluid inclusions studied represent the water-rich liquid.

A third possibility has been suggested by Weis. Our investigation has centered on the fluid inclusions. However, solid inclusions are also

present in the crystals of pegmatite minerals. Some are monomineralic and presumably represent accidental inclusions. Others, however, are of unknown composition; they may be mixtures of minerals. If so, have these mixtures crystallized from included mother liquor? At any rate, this matter needs thorough investigation. It is worth remarking that if minute disseminated fluid inclusions are always to be accepted as true samples of mother fluids, then the average granite crystallized from a water-rich solution.

By way of comment on the work of Roedder, it should be pointed out further that bulk analysis of fluid inclusions is valid as a guide to the composition of mineral-forming solutions only if it can be shown that the sole or at least predominant inclusions are of primary origin. In our work on ordinary pegmatite quartz, we have yet to find a specimen in which this is the case.

CONCLUSIONS

The following conclusions and comments appear indicated by the studies discussed above:

- (1) Inclusions in quartz and beryl in the pegmatites studied are of three principal types—(1) disseminated negative crystals or tubular inclusions, which in beryl are crystallographically oriented; (2) disseminated rounded or irregular inclusions; (3) inclusions along healed fractures. Inclusions of type (1) are probably primary, those of type (2) may be either primary or secondary, those of type (3) are secondary.
- (2) Inclusions of type (1) in the Selden east pegmatite and the Strickland-Cramer pegmatite consist of two phases at room temperature. These inclusions appear to be aqueous solutions with concentrations ranging from 2.7 to 6.5 molar. The vapor phases in various inclusions from the Selden east pegmatite disappear at temperatures ranging from 187° C. to 386° C. Pressure corrections equivalent to a rock load of 15,000 feet would raise these values to 272° C. to 600° C., a range for the possible temperature of crystallization that is unsatisfactorily broad. The range of values, uncorrected, is 196° to 268° C. for inclusions in beryl from the Strickland-Cramer pegmatite.
- (3) Inclusions of type (1) in beryl from six pegmatites consist of two liquid phases and a vapor phase at room temperature. One liquid phase disappears at about 31° C., hence appears to consist essentially of liquid carbon dioxide. With further heating, the vapor phase in some of the inclusions disappears, at 238° C. to 356° C. in various inclusions. In others the liquid phase dis-

- appears, at 310° C. to 455° C. In still others the liquid-vapor phase fades and disappears, at 329° C. to 348° C. Inclusions of all three types are present in beryl from three of the pegmatites.
- (4) Limited data suggest that inclusions of types (2) and (3) are comparable to those of type (1) in composition and behavior and hence were probably also formed during the period of pegmatite crystallization.
 - (5) Inclusions in quartz have proved unsatisfactory for temperature studies, because most of them fracture and leak when heated. Inclusions of type (1) are uncommon. Most show critical phenomena when heated and are of no value as indicators of temperatures of crystal formation. Most, perhaps all, such inclusions in quartz from the Case No. 2 pegmatite consist of three phases at room temperature. Inclusions of type (2) are more common. Those in quartz from the Case No. 2 pegmatite consist of three phases at room temperature, show disappearance of one liquid phase at about 31° C., and filling with liquid at 124° C. to 292° C. Inclusions of type (3) greatly predominate over the other types, but few data have been obtained, owing to fracture and leakage of the inclusions when heated.
 - (6) Decrepitation curves for quartz in pegmatites are significant only when bursting can be correlated precisely with the various types of inclusions, when the inclusions can be shown to consist of aqueous solutions, when premature bursting or leakage can be shown not to take place, and when the effects of primary inclusions are not masked by those of secondary inclusions. Our study suggests that these conditions will prove difficult to satisfy.
 - (7) Analysis of potential causes of variations in temperatures of disappearance of vapor and liquid phases and liquid-vapor boundaries indicates that a unique interpretation of the temperature data obtained is impossible at present. The actual temperatures of formation, uncorrected for pressure, may lie at one end or the other of the ranges of temperature values obtained, and there is no assurance that the statistical norm is significant.
 - (8) Absolute proof that inclusions in pegmatite minerals are primary is difficult to obtain, particularly for inclusions in quartz.
 - (9) True temperatures of pegmatite crystallization are probably considerably higher than temperatures of phase or phase boundary disappearance in liquid inclusions, but until some unequivocal means of determining pressures at time of crystallization can be devised, estimated corrections for pressures are little more than guesses based on geological possibilities. Owing to this, to

uncertainties as to the compositions of liquid inclusions, and to lack of *P-V-T* data for systems other than water at high temperatures and pressures, the liquid inclusion method is not at present suited to accurate determination of temperatures of mineral formation in the pegmatites studied.

- (10) Several questions must be answered before inclusions in pegmatite minerals can be accepted as true samples of the liquids from which pegmatites crystallize.

REFERENCES

- BAILEY, S. W., and CAMERON, E. N. (1951), Temperatures of mineral formation in bottom-run lead-zinc deposits of the upper Mississippi Valley, as indicated by liquid inclusions: *Econ. Geol.*, **46**, 626-651.
- BOWEN, N. L. (1928), Geologic thermometry, in laboratory investigation of the ores. McGraw-Hill Book Co., New York, pp. 172-199.
- BOWEN, N. L., The broader story of magmatic differentiation, briefly told, in ore deposits of the western states: *A.I.M.M.E.*, New York, 106-128.
- BUERGER, M. J. (1932a), The significance of block structure in crystals: *Am. Mineral.*, **17**, 177-191.
- BUERGER, M. J. (1932b), The negative crystal cavities of certain galena and their brine content: *Am. Mineral.*, **17**, 228-233.
- BUNN, C. W. (1949), Crystal growth from solution. II. Concentration gradients and the rates of growth of crystals (Discussion): *Faraday Soc.*, no. 5, 132-144.
- CAMERON, E. N., JAHNS, R. H., MCNAIR, A. H., AND PAGE, L. R. (1949), The internal structure of granitic pegmatites: *Econ. Geol., Mon.* **2**, 115 pp.
- CAMERON, E. N., LARRABEE, D. M., MCNAIR, A. H., PAGE, J. J., SHAININ, V. E., STEWART, G. W., Pegmatite investigations in Maine, New Hampshire, and Connecticut: *U. S. Geol. Survey, Prof. Paper*, in preparation.
- CAMERON, E. N., ROWE, R. B., AND WEIS, P. L., Fluid inclusions in beryl and quartz from pegmatites of the Middletown district, Connecticut: *Am. Mineral.*, **36**, 906-910.
- CAMERON, E. N., AND SHAININ, V. E. (1947), Beryl resources of Connecticut: *Econ. Geol.*, **42**, 353-367.
- DREYER, R. M., GARRELS, R. M., AND HOWLAND, A. L. (1949), Liquid inclusions in halite as a guide to geologic thermometry: *Am. Mineral.*, **34**, 26-34.
- FOYE, W. G., AND LANE, A. C. (1934), Correlations by radioactive minerals in the metamorphic rocks of southern New England: *Am. Jour. Sci.*, 5th ser., **28**, 127-138.
- GREGORY, H. E., AND ROBINSON, H. H. (1907), Preliminary geological map of Connecticut: *Conn. Geol. and Nat. Hist. Survey, Bull.* **7**, 31 pp.
- INGERSON, E. (1947), Liquid inclusions in geologic thermometry: *Am. Mineral.* **32**, 375-388.
- JENKS, W. F. (1935), Pegmatites at Collins Hill, Portland, Connecticut: *Am. Jour. Sci.*, 5th series, **30**, 177-197.
- JOHNSON, D. W. (1931), Stream sculpture on the Atlantic slope: Columbia University Press, New York, 142 pp.
- KENNEDY, G. C. (1950), "Pneumatolysis" and the liquid inclusion method of geologic thermometry: *Econ. Geol.*, **45**, 533-547.
- LAEMMLEIN, G. (1929), Sekundäre Flüssigkeitseinschlüsse im Mineralien: *Zeit. Krist.*, **71**, 237-256.

- NEUMANN, H. (1948), On hydrothermal differentiation: *Econ. Geol.*, **43**, 77-83.
- PEACH, P. A. (1949), Liquid inclusions in geothermometry: *Am. Mineral.*, **34**, 460-461.
- PEACH, P. A. (1951), Geothermometry of some pegmatite minerals of Hybla, Ontario: *Jour. Geol.*, **59**, 32-38.
- ROEDDER, E. (1951), Composition and significance of fluid inclusions in minerals: *Geol. Soc. Am., Bull.*, **62**, 1472, and oral presentation *Geol. Soc. Am.*, Annual meeting, Detroit, November, 1951.
- SCOTT, H. S. (1948), The decrepitation method as applied to minerals with fluid inclusions: *Econ. Geol.*, **44**, 465-484.
- SMITH, F. G. (1948*a*), The ore deposition temperature and pressure at the McIntyre mine, Ontario: *Econ. Geol.*, **43**, 627-636.
- SMITH, F. G. (1948*b*), Transport and deposition of the nonsulphide minerals. III. Phase relations at the pegmatitic stage: *Econ. Geol.*, **43**, 535-546.
- SMITH, F. G. (1949), Laboratory testing of "pneumatolytic" deposits: *Econ. Geol.*, **44**, 624-625.
- SMITH, F. G., *et. al.* (1950), "Pneumatolysis" and the liquid inclusion method of geologic thermometry (a reply to G. C. Kennedy): *Econ. Geol.*, **45**, 582-587.
- TWENHOFEL, W. S. (1947), The temperature of crystallization of a fluorite crystal from Luna County, New Mexico: *Econ. Geol.*, **42**, 78-82.

Manuscript received June 12, 1952.