THE SYSTEM, $Na_2O.Al_2O_3.2SiO_2$ (NEPHELITE, CARNEGIEITE)—$Na_2O.Al_2O_3.6SiO_2$ (ALBITE).

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ABSTRACT.

The results of an experimental study of equilibrium in the system, $Na_2O.Al_2O_3.2SiO_2$ — $Na_2O.Al_2O_3.6SiO_2$, are summarized in an equilibrium diagram, Fig. 1, and some of the relations to other systems are indicated in another diagram, Fig. 2.

INTRODUCTION.

This paper presents the results of an experimental investigation of phase equilibrium relationships in the system, $Na_2O.Al_2O_3.2SiO_2$ (nephelite, carnegieite)—$Na_2O.Al_2O_3.6SiO_2$ (albite), at atmospheric pressure, over a range of temperature of about 500° C., within which all the melting phenomena occur. The results are summarized and shown graphically in an equilibrium diagram, Fig. 1, and the principal data, on which the diagram is based, are collected in tabular form.[1] The relationships in this system, with albite as one end member, are similar to those found in the corresponding system, in which

[1] In this paper, temperatures are expressed in terms of the scale established by Day and Sosman (Day, A. L., Sosman, R. B., and Allen, E. T.: Carnegie Inst. Washington, Publ. No. 157, 1911) using the curve fitted to their data by Adams (Adams, L. H.: J. Am. Chem. Soc., 36, 65-72, 1914). The scale differs from the "International Temperature Scale." Several estimates of the differences between the two scales have been published, e.g. Wensel, H. T., and Roeser, W. F.: Bur. Standards Journal of Research, 5, 1309-1318, 1930; Sosman, R. B.: J. Am. Ceram. Soc., 16, 54-57, 1933. In the temperature region with which this paper deals, values of temperatures expressed in terms of the "International Temperature Scale" are probably always slightly higher than values for the same temperatures expressed in terms of the scale of Day and Sosman.

the place of albite is taken by anorthite, the other end member of the plagioclase series.[2]

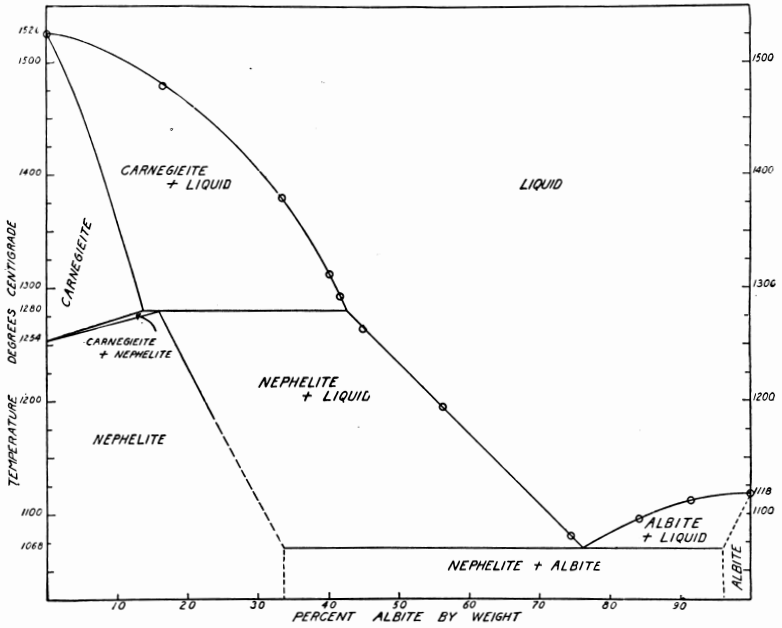

Fig. 1. Equilibrium diagram for the system, $Na_2O.Al_2O_3.2SiO_2$ (nephelite, carnegieite)—$Na_2O.Al_2O_3.6SiO_2$ (albite). The circles indicate determination of points on the liquidus. For other data see tables.

No discussion of the relationships shown on the equilibrium diagram seems called for, as similar systems have been published before, and no discussion is given here. In the pages that follow, however, a number of points in the experimental work are discussed in more or less detail.

EXPERIMENTAL.

Preparation of Glasses.

The first step in the experimental work was the preparation of a series of homogeneous glasses. The raw materials used were:

Silica, a crushed quartz, which after evaporation with hydrofluoric and sulphuric acids leaves a residue of less than 0.04 per cent, was converted to cristobalite by firing it at a high temperature.

[2] Bowen, N. L.: This Journal, 33, 551-573, 1912.

Alumina, prepared from the distilled chloride by a double precipitation with hydrochloric acid gas and ignition of the resulting aluminum chloride six hydrate.

Soda, weighed as the carbonate. The source was a particularly good sodium carbonate monohydrate. An analysis by E. G. Zies gave: SO_3 <0.001 per cent, Cl <0.001 per cent, SiO_2 <0.002 per cent, $Al_2O_3 + Fe_2O_3$ 0.003 per cent, CaO 0.008 per cent, MgO none.

Instead of making up each glass directly from these materials, a preparation, containing about 35 per cent soda 65 per cent silica, was made up and used as a source of soda in preparing the ternary glasses, the purpose being to avoid, as far as practicable, loss of soda during the melting of the glasses. This preparation was made in an electric furnace with a maximum temperature of 1050° C. At this temperature the liquid retains a good deal of gas.[3] This was driven off by crystallizing the material. Careful weighing showed that after the gas had been driven off, and before the charge was crushed, the weight was that of the soda and silica originally put into the charge. Several analyses were made on these materials, and they agreed with the compositions as determined by synthesis. There was then no detectable loss of soda.

In preparing the ternary glasses some soda is volatilized from the more sodic glasses while they are being heated at high temperatures. In order to keep track of the composition, careful weighings were made to detect any changes of weight that might occur while the preparations were in the furnace. These glasses take up some moisture from the air during crushing.[4] After crushing, the charge was heated in an electric furnace in air at 900°, and any loss taken to be water. Subsequent loss, at higher temperatures, was taken to be soda. With increase in silica the loss during heating decreases. Charges of 10 grams of albite glass showed no loss after being heated for 3 hours at 1650°-1675°.

In general, the compositions of these glasses do not lie exactly in the binary system, but, neglecting impurities, are expressible in terms of Na_2O, Al_2O_3, and SiO_2. The composi-

[3] This material, after being prepared in the electric furnace in air, increases in weight if heated over a meker burner (900° — 1000° C.), presumably by dissolving gases from the flame.

[4] For an interesting series of data on the moisture taken up by powdered feldspar crystals and glasses see A. L. Day and E. T. Allen: Isomorphism and Thermal Properties of the Feldspars, p. 57, Carnegie Inst. Washington, Publ. No. 31, 1905.

tions were plotted on a ternary plot, and from the points representing the compositions perpendiculars were drawn to the binary join. The intersections of these perpendiculars with the binary join give the compositions stated in the tables. These perpendiculars are, in all cases, so short that no significant error could be introduced into the liquidus relationships by the indicated departure of the composition from the binary system. The chief effect is likely to be the persistence of some residual liquid when an attempt is made to crystallize a mixture completely.

Melting Temperature of Albite.

The investigation of silicate equilibria dates properly from the work of Day and Allen on the feldspars.[5] In that work they began their melting determinations on anorthite. By making heating curves on the crystalline material they were able to determine the melting point. But, as they extended their investigation to more albitic feldspars, they found that the discontinuity on the heating curves was spread out to such an extent that the method gave no well defined melting temperature, and before albite was reached, gave no results at all. This led them to study the character of the change from the crystalline to the liquid state in the alkali feldspars. By holding the material at a constant temperature, for a long period of time, then cooling it, and examining the product with the microscope, they were led to recognize, for the first time, the existence of materials that can persist, in the crystalline state, for considerable periods of time at temperatures far above their melting temperatures, and that melt with extreme slowness in the lower portion of this range of instability. These observations have been abundantly confirmed since that time, and experience with silicates leads to the belief that the persistence in the crystalline state at temperatures above the melting temperature is a general property of silicates, and probably of matter, the differences being of degree.

Day and Allen found traces of melting in natural albite as low as 1100°, but were not able to establish a melting temperature. They say (op. cit., p. 50): "We did not extend any single trial beyond a single day, so that our results can not pretend to establish the lowest point at which albite melts.

[5] Op. cit.

Such an effort with a natural specimen known to contain impurities would yield nothing of value."

Bowen,[6] some years later, made a study of the plagioclase feldspars, determining the melting intervals of the series and drawing up an equilibrium diagram. Like Day and Allen, he was unable to crystallize albite, so used natural material. He found traces of glass after one hour at 1105° and no change after 2 hours at 1089°. He placed the temperature of the solidus for this material at 1100° ± 10° and the melting point of pure albite at the same temperature. We have not repeated this work on natural albite, our experiments being confined to synthetic material. The value of the melting point that we obtained is higher than this; 1118°, and the sluggish nature of the reactions in our material leads to the suspicion that the material that Bowen had was inhomogeneous.

The melting temperature of a crystalline substance, that melts congruently, is the temperature at which it can coexist in equilibrium with a liquid of the same composition. Above this temperature, the equilibrium state is one of complete liquidity, below it, of complete crystallinity. With some substances these reactions proceed so rapidly that they are not heated significantly above their melting temperatures without melting. In such cases failure to melt at a given temperature is sufficient indication that the temperature is below the melting temperature. However, with materials like albite, that melt to very viscous liquids, though melting at a given temperature shows that that temperature is above the melting temperature, failure to obtain melting does not show that it is not. Growth of crystals, on the other hand, does show that the temperature at which the growth occurs is below the melting temperature. We have fixed the melting temperature of pure albite, within limits, by finding a temperature at which crystals of albite melt to an albite liquid, and a second temperature at which they grow in an albite liquid. This was done by first growing small crystals of albite in a homogeneous glass of albite composition. The material, so prepared, was then held for some time at a constant temperature, then, by using the microscope, compared with the original material. If necessary, the heating time was increased until there was an unmistakable change in the size of the albite crystals. In this way a bracket was established. Then, by making longer runs, the bracket was

[6] Bowen, N. L.: This Journal, 35, 577-599, 1913.

TABLE I.

Melting points of albite.

Temp. °C.	Original condition	Time	Result
1113°	Crystals in glass	3 days	Crystals have grown
	Crystals	3 days	No change
1116°	Crystals in glass	2 days	Can see no change
1122°	Crystals in glass from run at 1113° above	1 day	Can see no change
		2 days	Crystals are smaller
		3 days	Nearly all glass
	Crystals	1 day	A little interstitial glass
		2 days	Glass increasing
	A bracket 1113°-1122° is established by these runs.		
1126°	Crystals in glass	1 day	All glass
	Crystals	1 day	Films of glass between the crystals throughout the charge
		2 days	Charge now contains only a few per cent of crystals. The smaller crystals have melted.
		3 days	Crystals have decreased
		4 days	Traces only of crystals
1115°	Crystals in glass	7 days	Crystals have grown
1120°	Crystals in glass	7 days	All glass

The melting temperature lies between 1115° and 1120°, probably at 1118°.

narrowed.[7] The results of these runs are listed in Table I, and the final bracket appears again in Table II.

The preparation of material suitable for these runs required considerable experimentation. We grew crystals of albite in the powdered albite glass in air but did not use them for the final work, for we found that by growing them under a small pressure of water vapor in sealed tubes of silica glass, we had a much better choice of size and distribution of crystals. In the same way we also prepared crystalline albite free from all but small traces of glass. Powdered albite glass was placed in a crucible of platinum foil, and the platinum folded down tightly over the charge, sealing it in. The charge, together with a source of water, was then sealed in an evacuated tube of silica glass and heated in an electric furnace. As a source of water we used gibbsite in some cases, and a rhyolitic glass

[7] These runs were possible only through automatic controlling and recording of the temperature. The control was by means of a regulator of the type due to White and Adams (Phys. Rev., 14, 44-48, 1919) and adapted by Roberts to use on alternating current. See especially J. Opt. Soc. Amer. and Rev. Sci. Instr., 11, 171-186, 1925. The recording potentiometer was built for this Laboratory by Leeds and Northrup.

TABLE II. Liquidus of carnegieite, of nephelite, and of albite.

Per cent albite by weight	Temp. °C.	Time	Original condition	Condition after runs
0.0	1524	30 min.	Glass	Glass + some carnegieite
			Carnegieite	Carnegieite
0.0	1527	30 min.	Glass	Glass
			Carnegieite	Glass with trace of carnegieite
		60 min.	Carnegieite	Glass
		The melting point is closer to 1527° than to 1524°, therefore at 1526°		
16.0	1476	60 min.	Glass	Glass with carnegieite
	1480	60 min.	Glass	Glass
			Glass + carnegieite	Glass
33.5	1374	60 min.	Glass	Glass + carnegieite
			Glass + carnegieite	Glass + carnegieite
		60 min.	Glass + carnegieite	Glass
40.2	1307	120 min.	Glass + carnegieite	Glass + carnegieite
			Glass	Glass + carnegieite
	1316	75 min.	Glass + carnegieite	Glass
			Glass	Glass
41.7	1289	120 min.	Glass	Glass + carnegieite
	1297	120 min.	Glass	Glass + trace carnegieite
			Glass + nephelite	Glass + trace carnegieite
45.1	1256	150 min.	Glass + nephelite	Nephelite crystals grown
	1260	150 min.	Glass + nephelite	No definite change
	1264	150 min.	Glass + nephelite	Glass
56.4	1190	17 hrs.	Glass + nephelite	Nephelite crystals grown larger
		5 hrs.	Glass + nephelite	Nephelite almost all gone
74.3	1072	48 hrs.	Glass + nephelite	Nephelite has grown
	1080	48 hrs.	Glass + nephelite	Can see no change
	1089	24 hrs.	Glass + nephelite	Nephelite all gone
84.2	1088	72 hrs.	Glass + albite	Albite grown
	1097	48 hrs.	Glass + albite	Albite slightly smaller
	1101	48 hrs.	Glass + albite	All glass
91.6	1102°	24 hrs.	Glass + albite	Albite grown
	1110	96 hrs.	Glass + albite	No change seen
	1115	19 hrs.	Glass + albite	Albite nearly all melted
100.0	1115+	168 hrs.	Glass + albite	Albite crystals grew
	1120—	168 hrs.	Glass + albite	All glass

containing about 3 per cent of water in others. Under these conditions some volatilization of silica from the walls of the tube occurs, but there is no detectable contamination of the charge inside its platinum container.[8]

Before any of these materials were used for the equilibrium investigations, the finely powdered materials were heated in air, close to the melting temperature, for 24 to 48 hours to make sure that they were dry.

Merwin[9] had, some years before, found that some change takes place in natural albite crystals, when they are heated in the neighborhood of 900°, and that it takes place in a few hours. As the duration of our runs was always several times as great as this, we did not expect any difference in the behavior of crystals grown at different temperatures. However, we tried crystals grown at 800°, 930°, and 1000°, but found no differences in behavior.

It will be seen from the data of Table IV that crystals of albite in an albite liquid persist for long periods of time at temperatures above the melting point. Thus after four days at 1126°, some crystal remnants still persist. A microscopic examination of the charges, that were held at temperatures above the melting point, shows that, at these temperatures (no experiments were made at temperatures far above the melting point), the melting occurs at the surfaces of crystals. The crystals gradually become smaller in size. The length of time required to melt a charge of albite at a given temperature depends therefore on the size of the individual crystals. These crystals in contact with a liquid of their own composition behave exactly like the same crystals in a somewhat different liquid when held just above the temperature of the liquidus, e.g. the albite crystals in the 91.6 per cent liquid at 1115° (see Table I). This is in agreement with Day and Allen's observations on the natural crystals, that the melting took place along cleavages. It is of interest in this connection, that quartz, which, like albite, may persist far above its melting temperature,[10] likewise melts at the surface, at least over a considerable range of temperatures above the melting temperature.

[8] Greig, J. W., Merwin, H. E., and Shepherd, E. S.: This Journal, 25, 61-73, 1933.

[9] Merwin, H. E.: J. Wash. Acad. Sci., 1, 59-60, 1911.

[10] This was reported by Ferguson and Merwin (This Journal, 46, 417-426, 1918) and has been repeatedly observed by one of us. An extreme example may be cited. A particularly fine quartz crystal was heated to about 1800° C. in an effort to melt it quickly. After cooling, it was found that the outer portion was silica glass but the inner part was still crystalline quartz.

Melting Temperature of Carnegieite.

The melting temperature of albite was located by showing it to be below one temperature, at which the crystals melted, and above another temperature, at which crystals grew. The melting temperature of carnegieite has also been bracketed, but, that the lower of the two temperatures is below the melting temperature, is shown in a somewhat different way. If a charge of powdered glass, of the composition of carnegieite, weighing a few hundredths of a gram,[11] is plunged into a furnace, already at the melting temperature of carnegieite, and is quenched shortly before the charge reaches this temperature, it will be found, on microscopic examination, that there is no visible glass. The crystallization, however, is spherulitic, and presumably there is interstitial glass. If, instead of quenching such a charge, it is held just under the melting temperature, or, if the quenched charge is returned to the furnace, and held at that temperature, liquid will collect, and large solid crystals of carnegieite will grow in it. The charge, originally glass, of the first run listed in Table II is an example.

Although the rate of approach to equilibrium, when carnegieite is heated just above its melting temperature, is slow enough so that it can be held there for an appreciable time without melting completely, it is in marked contrast with the rate at which equilibrium is approached when albite is so treated. So rapid indeed is this approach that Bowen[12] was able to determine the melting point by making heating curves, and the temperature so determined agrees with the temperature determined by the runs listed in Table I. No doubt this *exact* agreement is fortuitous, but it serves to illustrate the remarkable difference in behavior of carnegieite and albite near their melting temperatures—a difference very similar to that found by Day and Allen between the behaviors of anorthite and albite.

Liquidi of Albite, Nephelite, and Carnegieite.

A point on the liquidus of a crystalline phase represents the temperature and composition of a liquid that can coexist in

[11] A typical charge might weigh 0.035 grams and be held in a flat envelope of platinum foil weighing about 0.06 grams and measuring 6 x 8 mm.

[12] Bowen, N. L.: This Journal, 33, 551-573, 1912. Two heating curves gave 1525° and 1527° respectively; he therefore placed the melting point at 1526°.

equilibrium with the crystals. Experiments determining two such points, the melting points of albite and of carnegieite, have been described above. The growth or melting of crystals, in the experiments by which these special points were located, is a simpler process than it is in similar experiments, made on materials of intermediate compositions. In the general case, in which melting at equilibrium takes place, not at one temperature, but over a range of temperatures, the crystals grown in the liquid, preparatory to the determination of a point on a liquidus, may or may not be of the same composition as the liquid itself, but the composition of the crystals, that can coexist in equilibrium with the liquid, is always different from that of the liquid. The total mass of the small crystals, that are developed in the liquid, in order to make the determination, is so small that their formation changes the total composition of the remaining liquid by an amount that is equal to but a small fraction of the uncertainty in the composition of the liquid. This change in total composition of the liquid is therefore negligible. However, it is not the bulk composition of the liquid, but the composition of that liquid immediately surrounding the crystals at any given time, that determines the direction of change in the crystals. An analysis of the different possibilities shows that some melting of these crystals may take place at a temperature below that of the liquidus, but that complete melting cannot do so. On the other hand, provided the material has not been heated at a temperature above that of the liquidus, growth of the crystals can only take place at temperatures below that of the liquidus. The temperature of the liquidus, for any given composition, may then be bracketed by finding a temperature, at which crystals, grown in an originally homogeneous liquid of that composition, are completely melted, and another temperature at which they increase in size.

The data of the principal runs used in determining the temperatures corresponding to points on the liquidus of each of the three crystalline phases albite, nephelite, and carnegieite are listed in Table II. In each case appropriate measures were taken to ensure that suitable crystals were present in the charges when they reached the temperature of the run. With the exception of the preparations containing 74.3, 84.2, and 91.6 per cent albite, this presented no difficulty. In these, however, the development of crystals was slow, and although we grew crystals in all these glasses in the dry way, the material

actually used to locate the liquidus at 91.6 per cent albite was prepared with the aid of sealed tubes.

The Eutectic.

The composition of the eutectic, 76 per cent albite, is given by the intersection of the liquidus of albite with that of nephelite. This intersection also gives the temperature. Inde-

TABLE III.

Eutectic temperature.

Per cent albite by weight	Temp. °C.	Time	Original condition	Condition after runs
74.3	1063	48 hrs.	Crystalline	No sign of melting
	1072	24 hrs.	Crystalline	Mostly glass
84.2	1063	48 hrs.	Crystalline	No sign of melting
	1072	24 hrs.	Crystalline	Good deal of glass

pendent evidence on the temperature was obtained by finding a temperature at which melting occurred in originally crystalline charges of the two preparations closest to the eutectic in composition, and another temperature at which it did not occur. The crystalline material was prepared in a sealed tube, and, in order that the runs at constant temperature might be as short as possible, fine-grained material was selected. The results of these runs are listed in Table III. In this case the equilibrium condition has not been approached by growing crystals, so these runs do not demonstrate that the temperature 1063°, at which no melting was obtained in 48 hours, is below the temperature of the eutectic. The judgment that it is below rests on the contrast between the product of this run at 1063° and the product of the run at 1072°. It has been placed at $1068° \pm 5°$.

Solidi.

The solidus of a crystal species is a curve, all points on which represent the temperature and composition of crystals of the given species that can coexist in equilibrium with liquid. Thus in the equilibrium diagram, Fig. 1, the curve separating the field labelled carnegieite from that labelled carnegieite + liquid is the solidus of carnegieite. A downward extension of this curve is probably realizable in practise, but the conditions represented by it would be metastable, and the diagram represents

TABLE IV.

Solidus of carnegieite and of nephelite.

Per cent albite by weight	Temp. °C.	Time	Original condition	Condition after runs
5.2	1459	13 hrs.	Carnegieite	Carnegieite with a little interstitial glass
	1447	21 hrs.	Glass	Carnegieite. No glass found
10.9	1439	9 hrs.	Carnegieite	Carnegieite with interstitial glass
15.0	1288	15 hrs.	Glass	Carnegieite with traces of glass
15.8	1317	16 hrs.	Carnegieite	Carnegieite with glass
17.0	1288	45 hrs.	Glass	Carnegieite with glass
17.0	1268	21 hrs.	Nephelite	Nephelite + glass
	1259	23 hrs.	Nephelite	Nephelite + trace of glass in rare grains
	1250	45 hrs.	Nephelite	Nephelite. No glass found
20.0	1268	21 hrs.	Nephelite	Nephelite + glass
	1259	23 hrs.	Nephelite	Nephelite + glass
	1250	45 hrs.	Nephelite	Nephelite + glass
	1226	42 hrs.	Nephelite	Nephelite. No glass found
25.3	1200	24 hrs.	Nephelite	Nephelite + glass
			Glass	Nephelite + glass
	1160	48 hrs.	Nephelite	Nephelite
			Glass	Nephelite

only stable equilibrium. The composition of the liquid, with which the crystals can coexist in equilibrium at any temperature, is shown by a corresponding point on the liquidus.

The data used in locating the solidus of carnegieite and that of nephelite are given in Table IV. The data on the solidus of albite are in the text of the latter part of this section. As will appear from the following discussion, these curves are not located as well as the corresponding liquidi.

The experimental work to locate the solidus of carnegieite, and that of nephelite, aimed, first, at the complete crystallization of a series of homogeneous glasses to crystals of uniform composition, and then at the determination of the lowest temperature at which liquid would form from these crystals. The decision that a temperature is not above that of the solidus rests on the non-occurrence of melting at the given temperature. With very viscous materials this, by itself, would prevent the location of a point on the solidus with the same satisfaction as a point on the liquidus at the same temperature.

The preparation of completely crystalline material is frequently difficult. Indeed one rarely succeeds in *completely* crystallizing a silicate preparation. It is evident that, if the

composition of the glass is not exactly that of the crystals that separate from it, some residual liquid will remain. The failure of the glass to be of exactly the right composition may be due to any of a number of causes such as: impurities in the materials from which the glass is made, differential loss of materials in making the glass, inhomogeneity causing local departures from the correct composition, etc. Yet, when allowance is made for all possible departures of the glass from the right composition, it commonly seems as if there must be something more than this preventing the crystallization of the last interstitial liquid.

In general, then, the determination that a temperature is above that of the liquidus for a given composition involves judgment of the relative amount of glass in two preparations: the original crystallized material and the material that has been heated at the given temperature. The more completely crystallized the original material is, the more readily small increases in the amount of glass can be recognized, and, other things being equal, the closer to the solidus the temperature at which melting can be recognized.

The glasses used in the determination of points on the solidus of nephelite crystallized rapidly to a condition in which no trace of glass was visible. However, judging from the nature of the extinction, the crystallization was spherulitic, and experience with silicates has given ample reason to suspect that such material may be but partially crystallized. Complete crystallization usually results in the development of numerous voids,[13] and these products were quite free from voids. Moreover, the index of refraction was low. After the material had been heated for several days, voids were plentiful, and the index of refraction was up, but it was not possible to say whether or not the material was completely crystallized to nephelite, for the index of refraction is not sufficiently sensitive to small amounts of glass or of carnegieite, and the crystallization was still fibrous with no individual crystals to be seen. When these preparations were heated at higher temperatures, glass appeared, voids disappeared, and blocky crystals of nephelite developed, all at the same temperature. We have drawn the solidus below points representing the temperature at which this change took place, and above points representing temperatures at which it did not occur.

[13] Probably due to shrinkage, but in some cases possibly in part also to the concentration and separation of gas present in the original glass.

At temperatures toward that of the eutectic the above method was unsatisfactory. We crystallized glasses containing 25.3, 33.5, and 40.2 per cent albite below the eutectic temperature. The products were spherulitic, however. We then held these materials for 3 days at 1080°. No change occurred in the 25.3 per cent material, both the others were largely unchanged but locally traces of glass had developed in each of them. Doctor Posnjak kindly made comparison X-ray powder spectrograms for us, and these showed a slight but definite difference in the positions of the lines in the patterns given by the 33.5 and the 25.3 per cent materials, but no difference was observable between the patterns from the 33.5 and the 40.2 materials. This indicates that at this temperature solid solutions extend beyond 25 per cent, but does not fix the limit. We have drawn the solidus tentatively through 33 per cent at 1080°, but hope to be able to locate it more definitely.

While we were working with these mixtures we crystallized some of the glasses at 600° and Doctor Posnjak made an X-ray examination of them. The photographs agree with those made on the material described above, showing no difference in the extent of solid solution at that temperature.

The glasses used for locating the solidus of carnegieite also crystallized rapidly to a spherulitic mass, but on continued heating in the range of stability of carnegieite, solid blocky crystals of carnegieite developed, and the cooled charges consisted of an aggregate of these crystals showing the intricate twinning due to the inversion from the high-temperature isotropic form to the low-temperature birefringent form.[14] When liquid is present it collects in the interstices. The longer the heating the coarser are the crystals and the better is the opportunity to see a given amount of interstitial glass. Small amounts of interstitial glass can be seen only by virtue of a difference between the refractions of the glass and the crystals. The index of refraction of carnegieite is so close to that of the glass that it would be quite impossible to see the first traces of glass developed, and this difficulty is accentuated by the twinning.

A question of considerable interest to petrology is whether or not the composition of albite can vary toward nephelite, that is, whether or not solid solutions lower in silica than albite occur in this binary system. In an attempt to answer this, glasses containing 90, 95, 97, 98, and 100 per cent of albite

[14] Bowen, N. L., and Greig, J. W.: This Journal, 10, 204-212, 1925.

were prepared, then crystallized by heating in sealed tubes, followed by two months in air at about 1035°. Several preparations were made from each glass and the most favorable was selected. The crystals obtained in this way are not spherulitic, and small traces of interstitial glass are easily seen. It has already been said that it is usually difficult to crystallize a material *completely*, and these compositions are more difficult to crystallize than most that have been investigated. It was possible under the microscope to see glass in all these preparations; nevertheless they were excellently crystallized.

If it were possible to show whether or not nephelite was present in these preparations, one could, by the presence of nephelite, fix a limit to the extent of solid solution at the temperature at which crystallization took place, and by its absence, in these well-crystallized preparations, fix a composition to which solid solutions extend. However, this could not be done, either by the microscope or by the X-ray. The indices of refraction of nephelite and of albite are so nearly the same that small amounts of nephelite would not be seen in these fine-grained preparations, and Doctor Posnjak found, on comparing the spectrograms of albite and nephelite solid solutions, that the strong lines of each pattern were so nearly identical in position that it would be impossible by this means to detect a few per cent of nephelite crystals mixed with albite.

Although the expectable change in spacing accompanying a small change in the silica content of albite is very slight, Doctor Posnjak kindly made comparison X-ray powder spectrograms of these materials using molybdenum $K\alpha$ radiation. No differences were found.

The preparations were heated at 1072°-1075°, that is, just above the eutectic temperature, for three days, after which some glass was present in the 90 and 95 per cent preparations. After a further eight days at this temperature, examination showed more glass in the 90 per cent preparation but no apparent change in the 95 per cent. No detectable change took place in the 97 or 98 per cent preparation. Other charges of the same materials were heated for fifteen days at 1079° with the same results.

The evidence leaves much to be desired, but, on the basis of these experiments, we are of the opinion that some solid solution does occur, and tentatively have placed the limit at 1075° between 95 and 97 per cent albite, i.e. at 96 per cent albite. We hope to secure further information on this point.

The Nephelite-Carnegieite Inversion.

The principal data fixing the temperature of this inversion are listed in Table V. To determine these temperatures we crystallized samples of the glasses (with the exception of the 33.5 per cent glass) completely to carnegieite and to nephelite. No difficulty was experienced in obtaining materials completely carnegieite, but we found that in the temperature region in which crystallization is fairly rapid, carnegieite formed first and that nephelite was obtained only by inverting the carnegieite. To convert the carnegieite completely to nephelite was a slow process. Instead of doing this, charges were crystallized to nephelite at low temperatures in sealed tubes. The inversion temperature, in the pure end member, was found to lie between 1259° and 1249°. Bowen[15] found it to lie between 1245° and 1252°.

With increase in the silica content of the solid solutions the temperature of the inversion rises. The change does not then take place at a single temperature but over a range of temperatures, as indicated on the equilibrium diagram by the field labelled carnegieite + nephelite. The data do not well define the width of this field. Were it possible to locate the solidus curves of nephelite and of carnegieite more closely the boundaries would of course be better located.

Jadeite.

The composition of jadeite, $Na_2O.Al_2O_3.4SiO_2$, lies between that of nephelite and albite, but no crystals of jadeite were found in any of our preparations. The non-occurrence of crystals of this compound does not prove that it is not stable. However, if crystals of this compound decompose under these conditions, the compound is unstable, and does not enter into the equilibrium diagram. In an effort to decide this, we obtained several specimens of jadeite, and, on the basis of microscopic examination, selected one of them. This was a piece from a specimen of white jadeite from Burma, U. S. National Museum number 94303, which we obtained through the courtesy of Dr. William F. Foshag. The specimen contains good-sized crystals of jadeite mixed with a little interstitial albite. The albite was removed by using a heavy liquid.

[15] Bowen, N. L.: This Journal, 33, 551-573, 1912. It may be noted in passing that whereas our materials inverted very slowly indeed, Bowen obtained almost complete conversion of nephelite to carnegieite in one hour at 1252°, and partial conversion of carnegieite to nephelite in one hour at 1245°.

TABLE V.

Nephelite-carnegieite inversion.

Per cent albite by weight	Temp. °C.	Time	Original condition	Condition after runs
0.3	1249	24 hrs.	Nephelite	Nephelite
			Carnegieite	Carnegieite
		48 hrs.	Nephelite	Nephelite
			Carnegieite	Carnegieite with a little nephelite
	1268	23 hrs.	Nephelite	Carnegieite
	1259	24 hrs.	Nephelite	Nephelite
		48 hrs.	Nephelite	Nephelite + carnegieite
5.2	1250	28 hrs.	Nephelite	Nephelite
			Carnegieite	Carnegieite + a little nephelite
	1259	48 hrs.	Nephelite	Nephelite
			Carnegieite	Carnegieite
	1269	24 hrs.	Nephelite	Nephelite
			Carnegieite	Carnegieite
	1277	24 hrs.	Nephelite	Nephelite
			Carnegieite	Carnegieite
		48 hrs.	Nephelite	Nephelite + carnegieite
	1285	4 hrs.	Nephelite	Nephelite + carnegieite
10.9	1269	24 hrs.	Nephelite	Nephelite
			Carnegieite	Carnegieite + rare nephelite
		48 hrs.	Carnegieite	Carnegieite + increase in nephelite
	1277	24 hrs.	Nephelite	Nephelite
		48 hrs.	Nephelite	Nephelite + carnegieite
15.8	1259	48 hrs.	Nephelite	Nephelite + trace glass
	1269	24 hrs.	Carnegieite	Carnegieite + rare nephelite
		48 hrs.	Carnegieite	Carnegieite + increase in nephelite
	1273	22 hrs.	Carnegieite	Carnegieite + rare nephelite + trace glass
	1277	24 hrs.	Nephelite	Nephelite + trace glass
			Carnegieite	Carnegieite + trace glass
		48 hrs.	Nephelite	Nephelite + trace glass
	1285	24 hrs.	Nephelite	Nephelite + glass + a little carnegieite
33.5	1268	4 hrs.	Carnegieite + glass	Carnegieite + glass + nephelite
	1273	22 hrs.	Carnegieite + glass	Carnegieite + glass + nephelite
	1285	4 hrs.	Nephelite	Nephelite + glass
		24 hrs.	Nephelite	Nephelite + carnegieite + glass

The indices of the jadeite, for which we are indebted to Dr. H. E. Merwin, follow: β 1.657, the total variation of β being less than 0.002, γ 1.668 (highest found), α 1.650 (lowest found).

The jadeite was then heated in air in an electric furnace. Decomposition was observed as low as 800° C. After having been heated at 1015° it was entirely converted to glass and large nephelite crystals. Although this jadeite is not pure

$Na_2O.Al_2O_3.4SiO_2$, its conversion to nephelite plus liquid at higher temperatures is sufficient evidence to support the conclusion that jadeite is not stable under the conditions represented by the equilibrium diagram.

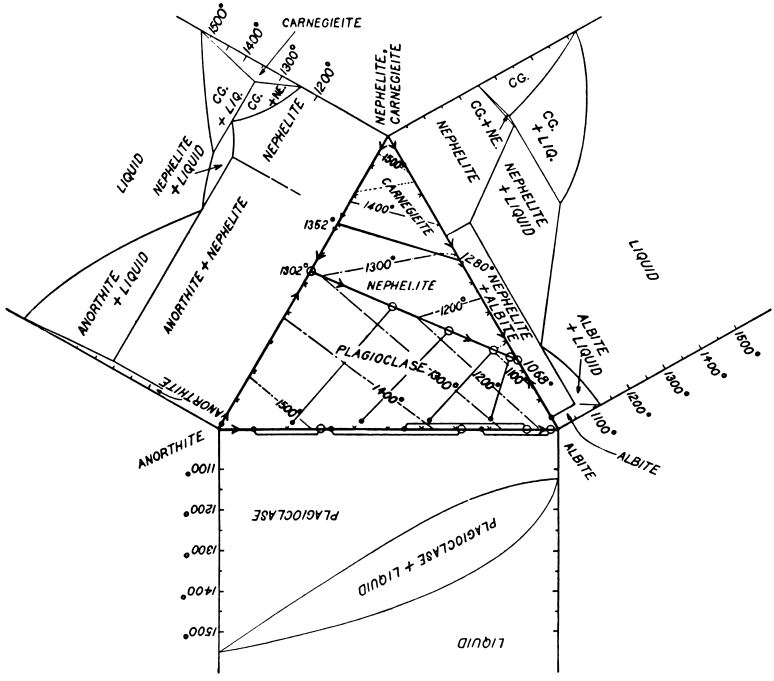

Fig. 2. The equilibrium diagrams for the systems, nephelite, carnegieite—anorthite (Bowen, N. L.: This Journal, 33, 551-573, 1912) and the binary system, albite—anorthite (Bowen, N. L.: This Journal, 35, 577-599, 1913), are combined with the diagram of Fig 1. The ternary diagram (which is entirely hypothetical) has been drawn from these three binary diagrams, points being located by straight line interpolation. An open circle represents the composition of a liquid that can coexist in equilibrium with crystals of the composition indicated by that filled circle that is joined to the open circle by a tie line.

SUMMARY.

The experimental work is summarized in the equilibrium diagram, Fig. 1. The limiting compositions of the solid solutions have not been fixed as well as we should like. It is hoped that it may be possible to obtain more information leading to a better determination of these relationships.

Although the composition of jadeite, $Na_2O.Al_2O_3.4SiO_2$, lies between that of nephelite and albite, it is evidently unstable under the conditions represented in the diagram. This was to be expected, for it is not known in the ordinary igneous rocks and has been considered by petrologists to be formed only at high pressures.[16, 17]

The equilibrium diagram for this system is very similar to that for the corresponding system, nephelite, carnegieite—anorthite, as determined by Bowen.[2] In both systems the compositions of the nephelite and the carnegieite vary within limits from $Na_2O.Al_2O_3.2SiO_2$ toward the composition of the feldspar, anorthite, or albite, or, expressing this variation in composition in other words, nephelite and carnegieite take a limited amount of feldspar into solid solution. In both systems the temperature of the inversion nephelite-carnegieite rises with increased feldspar content. At the other end of each diagram, as drawn, some slight solubility of nephelite, carnegieite in the feldspar is indicated.

Bowen found that with increasing anorthite content the birefringence of nephelite decreased, became zero, then increased again, the crystals having become optically positive. The small size of the nephelite crystals, that grew in the nephelite-albite liquids, prevented satisfactory measurements of the indices of refraction, but the change to optically positive nephelite does not occur in this system. These nephelites are all optically negative, as are the naturally occurring examples.

In view of the similarity of these two systems it is to be expected that closely similar relationships will be found to obtain between nephelite, carnegieite and feldspar across the ternary system, nephelite, carnegieite—albite—anorthite, modified of course by the more complex nature of the equilibrium in the ternary system. With the three binary systems known, and no intermediate compounds except jadeite known or to be expected, it is a simple matter to draw up a ternary diagram that, while not correct in detail, will show the general relationships. For the convenience of the reader such a diagram has been added (Fig. 2).

In Fig. 2 the equilibrium diagrams for the systems, nephelite, carnegieite—anorthite,[2] and albite—anorthite,[6] have been

[16] See P. Eskola: The mineral facies of rocks, Norsk Geol. Tid., 6, pt. 1-20, pp. 143-194, 1920 (The metamorphic eclogite facies, p. 168, the igneous eclogite facies, p. 182).

[17] Harker, A.: Metamorphism, London, 1932, p. 308.

reproduced and to them the diagram of Fig. 1 has been added. A ternary diagram has been drawn up by combining the data from these three. Positions of points within this diagram have been located by straight line interpolation. A number of tie lines have been indicated in the feldspar part of the diagram. These are lines joining the points representing the compositions of phases that can coexist in equilibrium with each other. The open circles represent liquid, the solid circles crystals. Tie lines within the diagram are of course hypothetical. It will be noted that one set of open circles lies apparently on the boundary between the fields of nephelite and of feldspar. Liquids represented by points actually on the field boundary are of course in equilibrium with both nephelite and feldspar. No tie lines have been drawn to the points representing the composition of the nephelite crystals. It may save confusion to suppose that these circles are very close to the field boundary but actually lie within the feldspar field. A thorough description of relationships of this kind has been given by Bowen[18] in his paper on the system, diopside—anorthite—albite.

[18] Bowen, N. L.: This Journal, 40, 161-185, 1915.