

THE MELTING OF ANALCITE SOLID SOLUTIONS
IN THE SYSTEM



TJ. PETERS,² W. C. LUTH³ AND O. F. TUTTLE,⁴

*Department of Geochemistry and Mineralogy,
The Pennsylvania State University,
University Park, Pennsylvania.*

ABSTRACT

Analcite melts incongruently to albite, nepheline and liquid above an invariant point at $4.75 \pm .25$ Kb and $665 \pm 5^\circ$ C. in the system $\text{NaAlSi}_3\text{O}_8\text{-NaAlSiO}_4\text{-H}_2\text{O}$. At the invariant point analcite has an anhydrous composition $\text{Ab}_{50}\text{Ne}_{50}$ and the liquid has an anhydrous composition $\text{Ab}_{53}\text{Ne}_{47}$, containing $15.5 \pm 0.5\%$ H_2O . The position of the five univariant reaction curves, meeting at the invariant point were determined between 1 and 10 Kb. The composition of the low melting liquid is enriched in nepheline and water with increasing pressure. At 6 Kb and 675° C. 5-10% solid phases are dissolved in the vapor phase. All five phases lie on the join $\text{NaAlSiO}_4\text{-NaAlSi}_3\text{O}_8\text{-H}_2\text{O}$. Infrared absorption proved to be a less sensitive method for evaluating the analcite composition than x-ray diffraction. Preliminary investigation of the effect of adding potassium to the system showed that analcite and liquid can coexist at 2.3 Kb and 650° C. Small amounts of potassium (about 2% K_2O) can be taken up by the analcite solid solutions.

It is suggested that analcite phenocrysts in some extrusive and hypabyssal rocks can form in the small temperature region where analcite coexists with liquid and that ground-mass analcite could crystallize from differentiates of alkali-rich magmas within the experimentally determined stability field of analcite.

INTRODUCTION

Analcite was long thought to be only a secondary mineral after sodalite or nepheline (Rosenbusch, 1908). Later the textural relations convinced most petrographers (Osann, 1923; Washington, 1914; Larsen and Buie, 1938; Lyons, 1944; Scott, 1916) that it could be a primary igneous mineral. The principal purpose of this study was to determine experimentally whether or not analcite is stable to high enough temperatures and pressures to coexist with magmatic liquids.

The PT curve for the decomposition of analcite according to the reaction $\text{analcite} = \text{nepheline} + \text{albite} + \text{V}$ extends to *higher* temperature as the pressure is increased and the Pt curve for the beginning of melting in the system nepheline-albite-water, according to the reaction $\text{nepheline} + \text{albite} + \text{vapor} = \text{liquid}$, extends to *lower* temperatures as the pressure is

¹ College of Mineral Industries Experiment Station. Contribution No. 65-7.

² Present Address: Mineralog.-Petrograph. Institute der Universität Bern, Sahlstrasse 6, 3012 Berne, Switzerland.

³ Present Address: Department of Geology and Geophysics, Massachusetts Institute of Technology, Cambridge, Massachusetts.

⁴ Present Address: Department of Geology, Stanford University, Stanford, California.

increased. The intersection of these two curves will generate an invariant point as described by Morey (1957). Furthermore, he demonstrated that the nature and position of the univariant curves extending to higher pressures is determined by the composition of analcite, liquid and vapor at the invariant point.

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EXPERIMENTAL METHOD

For the present study two sets of starting materials were used. One set consisted of most of the glasses used by Saha (1959). These glasses had been made partly by fusing gels, prepared by the method of Roy (1956) and partly by fusing different mixtures of $NaAlSiO_4$ and $NaAlSi_3O_8$ glasses. The second set consists of gels prepared by the method described by Luth and Ingamells (1965). These gels were made into glasses by repeated firing and grinding. The results obtained from both sets of glasses were in good agreement.

Weighed amounts of glass or crystalline material were sealed in gold capsules and run in cold-seal pressure vessels, described by Tuttle (1949) and Luth and Tuttle (1963). Pressures above 2 Kb were measured by a Harwood manganin cell, giving pressures reproducible to ± 0.2 Kb. A Honeywell-Brown circular scale indicating temperature controller was used to control the temperature which was measured by a Honeywell-Brown circular scale indicating potentiometer. The temperature is believed to be reproducible to within $\pm 5^\circ$ C.

The run products were examined with a petrographic microscope, x-ray diffraction and in some cases by infrared spectroscopy. For the latter a Perkin Elmer Model 21, using NaCl and KBr prisms and a Beckman DK2A spectrograph was used.

RESULTS

Crystalline Phases

a. *Analcite solid solutions* crystallize over a wide range of composition and temperature as has been shown by Barrer and White (1952) and

Saha (1961). The analcite crystals obtained in the runs were isotropic and had, under the microscope, almost circular outlines with a diameter of about 40μ . In one run clear icositetrahedra crystallized with a diameter of 1–2 mm. Saha (1959, 1961) showed that the index of refraction and the

TABLE 1. VALUES FOR $2\theta_{\text{An1}(639)} - 2\theta_{\text{Si}(331)}(\Delta)$ FOR Cu $K\alpha_1$
FOR DIFFERENT ANALCITE COMPOSITIONS

a) Values determined in the present study b) Values determined by Saha (1961)

a) Anhydrous Composition									
Mol. Ratio	Si	Ab	Ne	Δ in $^{\circ}2\theta$	Mol. Ratio	Si	Ab	Ne	Δ in $^{\circ}2\theta$
6.00*		Ab ₁₀₀	Ne ₀	2.18	3.74		Ab _{58.8}	Ne _{41.2}	1.72
5.41		Ab _{91.4}	Ne _{8.6}	2.08	3.50*		Ab _{52.6}	Ne _{47.4}	1.645
5.01		Ab _{84.9}	Ne _{15.1}	1.995	3.49		Ab _{52.2}	Ne _{47.8}	1.64
5.00*		Ab _{84.7}	Ne _{15.3}	1.99	3.25		Ab _{45.7}	Ne _{54.3}	1.585
4.32		Ab _{71.8}	Ne _{28.2}	1.83	3.25*		Ab _{45.6}	Ne _{54.4}	1.57
4.02		Ab _{65.3}	Ne _{34.7}	1.75	3.02		Ab _{39.2}	Ne _{60.8}	1.545
4.00*		Ab _{65.1}	Ne _{34.9}	1.80	3.00*		Ab _{38.1}	Ne _{61.9}	1.55
3.75*		Ab _{58.9}	Ne _{41.1}	1.71					

* Glasses used also by Saha (1961).

Standard deviation $\pm 0.03^{\circ}$.

Equation of the line $0.875 \pm 0.222X$.

Variance of slope $1.1 \cdot 10^{-5}$.

Variance of intercept $1.9 \cdot 10^{-4}$.

b) Anhydrous Composition									
Mol. Ratio	Si	Ab	Ne	Δ in $^{\circ}2\theta$	Mol. Ratio	Si	Ab	Ne	Δ in $^{\circ}2\theta$
6.00		Ab ₁₀₀	Ne ₀	2.123	3.50		Ab _{52.6}	Ne _{47.4}	1.680
5.00		Ab _{84.7}	Ne _{15.3}	1.940	3.25		Ab _{45.6}	Ne _{54.4}	1.610
4.00		Ab _{65.1}	Ne _{34.9}	1.763	3.00		Ab _{38.1}	Ne _{61.9}	1.540
3.75		Ab _{58.9}	Ne _{41.1}	1.770					

Standard deviation $+0.03^{\circ}$.

Equation of the line $1.020 + 0.185X$.

Variance of slope $1.1 \cdot 10^{-4}$.

Variance of intercept 2.10^{-3} .

cell edge of analcite solid solutions changes with the chemical composition. The variation in the cell edge can be used as a sensitive method to evaluate the chemical composition of the analcite. For this, the displacement of the Cu $K\alpha_1$ peak of the (639) reflection is measured against the Cu $K\alpha_1$ peak of the (331) reflection of a silicon standard. In Table 1, these values are given for analcites crystallized from the two different

sets of glasses, together with those obtained by Saha (1961). The equations for the best fitting lines for Saha's data and those of the present study were calculated for the analcite solid solutions between $\text{Na}_2\text{Al}_2\text{Si}_6\text{O}_{16} \cdot n\text{H}_2\text{O}$ and $\text{Na}_2\text{Al}_2\text{Si}_3\text{O}_{10} \cdot n\text{H}_2\text{O}$ (Fig. 1).

Analcite crystallized from compositions with less silica than $\text{Na}_2\text{Al}_2\text{Si}_3\text{O}_{10}$ showed no further displacement of the (639) reflection, although no phases in addition to analcite were found. The analcite lattice does not seem to be able to expand beyond this point. It is hardly a coinci-

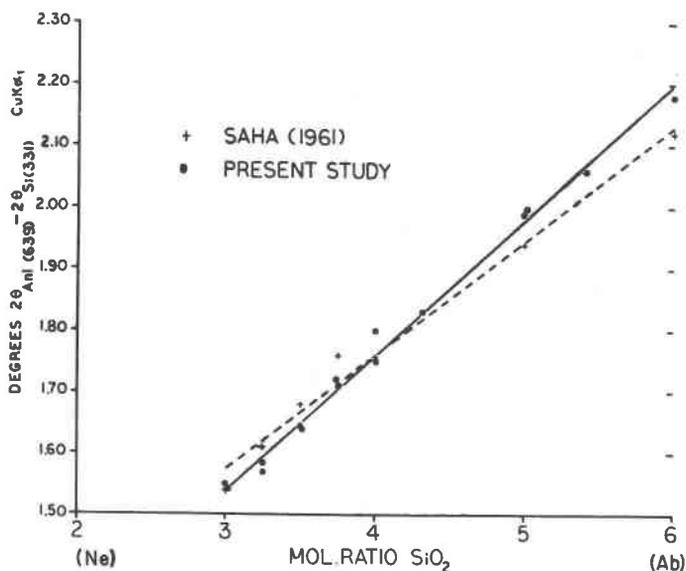


FIG. 1. Analcite determinative curve (after Saha, 1961) used in this study (Table 1).
Mol ratio SiO_2 = molecular ratio $\text{SiO}_2:\text{Na}_2\text{O}$.

dence that, in nature, at this composition, natrolite occurs. It has not yet been possible to synthesize natrolite.

Infrared spectra of analcite solid solutions were made to see how the spectra are affected by the $\text{NaAl}=\text{Si}$ substitution and to see if this method could eventually be used for the determination of the analcite composition. The absorption bands obtained from different analcite solid solutions are listed in Table 2. Most bands do not seem to be greatly affected by the changing analcite composition, but a few are: The band in the $9\text{--}10\mu$ region shifts toward longer wave lengths with decreasing Si content due to the substitution of Al for Si. In the 14μ region the Si-rich and Si-poor analcites have somewhat different bands. The band at 16μ also shifts toward longer wave lengths with decreasing Si content. These

results show that infrared spectroscopy is not as well suited for the determination of the analcite composition as x -ray diffraction.

b. *Nepheline solid solutions* commonly crystallize in short hexagonal prisms, but more commonly in fine grained aggregates and then could only be distinguished from albite by x -ray diffraction. The extent of nepheline solid solution in this system was determined just below and just above the invariant point at 5 Kb 645° C. and 6 Kb 670° C. respectively. Different H₂O contents were used to minimize the influence of loss to the vapor phase on the initial composition. The amount of solid solution of albite in nepheline at these pressures and temperatures lies between 10% and 15%.¹ This value is much lower than that obtained at

TABLE 2. INFRARED ABSORPTION BANDS FOR ANALCITES OF DIFFERENT COMPOSITIONS

Anhydrous Composition		Absorption—bands in μ									
		vs=very strong	s=strong	ms=medium strong		w=weak		br=broad			
Ab ₁₀₀	Ne ₀	2.75s	2.81m	6.15m	6.9w	br9.6vs	12.82s	13.6ms		15.9m	br22vs
Ab _{84.9}	Ne _{15.1}	2.76s	2.81m	6.15m	6.9w	br9.6vs	13.05ms	13.7ms		16.1m	br22vs
Ab _{71.5}	Ne _{28.2}	2.76s	2.81m	6.15m	6.9w	br9.7vs	13.2ms	13.7ms		16.1m	br22vs
Ab _{65.8}	Ne _{34.9}	2.77s	2.81m	6.15m	6.9w	br9.8vs	13.2ms	13.7ms		16.2m	br22vs
Ab _{58.9}	Ne _{41.1}	2.75s	2.81m	6.15m	6.9w	br9.8vs	13.2w	13.7ms	14.6w	16.2m	br22vs
Ab _{52.6}	Ne _{47.4}	2.78s	2.81m	6.15m	6.9w	br9.8vs		13.7s	14.7w	16.3m	br22vs
Ab _{46.6}	Ne _{53.4}	2.76s	2.81m	6.15m	6.9w	br9.8vs		13.6s	14.7m	16.35m	br22vs
Ab _{38.1}	Ne _{61.9}	2.77s	2.81m	6.15m	6.9w	br9.8vs		13.6s	14.7m	16.4m	br22vs
Ab _{26.1}	Ne _{73.9}	2.76s	2.81m	6.12m	6.9w	br9.8vs		13.6s	14.9m	16.5m	br22vs
Assignment of band		O-H	O-H	H ₂ O	Si-O						Si-O

1 Kb and the same temperature by Edgar (1964) where the amount of albite in nepheline solid solution is 20–25%. This suggests a considerable decrease of solid solution of albite in nepheline with increasing pressure.

To investigate the possibility of OH substituting for O in nepheline, as suggested by Smith and Tuttle (1957), glasses with different amounts of H₂O were crystallized into nepheline. Even with 1% H₂O the presence of a vapor phase could be detected. Infrared spectra of nepheline crystallized with and without H₂O at different pressures showed no OH-vibration bands besides the OH band for absorbed H₂O. This suggests that the OH substitution for O does not take place on a large scale in nepheline.

c. *Albite* crystallized always as high to intermediate albite (MacKenzie, 1957) and when not formed from the vapor, as fine needles. The amount of nepheline in solid solution is less than 5%; our composition closest to albite on the join.

¹ All percentages in this paper are in terms of weight per cent.

d. *Paragonite* rarely crystallized in the nepheline-rich part of the system at very high H₂O contents. Optically paragonite was difficult to distinguish from NaHCO₃, which often accompanies it in the products of our runs. Under the conditions used in the present study, paragonite formed in very small amounts and in only one run could be detected by x-ray diffraction.

e. *Nepheline hydrate I* was only encountered in the low pressure and low temperature crystallization runs, using compositions between Ne₁₀₀Ab₀ and Ne_{86.9}Ab_{13.1}.

f. *Phases crystallized from the vapor*. Considerable amounts of solid material are dissolved in the vapor phase, especially at higher pressures (Fig. 4). On quenching the dissolved material precipitates from the fluid in one of the following ways:

a) small spheres 10–50 μ are formed, that are isotropic and seem to consist of a glass or gel; b) crystals grow on the quench and are seen to be swimming in the juice on opening the capsules. These crystals can easily be distinguished from those that do not grow from the vapor, because they are bigger, idiomorphic and not associated with other crystals or glass; c) the third type grows after the capsules are opened and the aqueous fluid comes in contact with the atmosphere. In this case the carbonates appear.

The following crystalline phases originating from the vapor phase could be detected optically and by x-ray diffraction: Albite in big idiomorphic single twinned crystals, α -Al₂O₃ (corundum) in 40–100 μ hexagonal plates, paragonite, NaHCO₃ in highly birefringent needles, often twinned, and Na₂CO₃-H₂O in birefringent radial aggregates.

INVARIANT AND UNIVARIANT EQUILIBRIA

The pressure-temperature projection given in Fig. 2 is based principally on the experimental data given in Table 3. The invariant point where the five phases, analcite solid solution, nepheline solid solution, albite, liquid, and vapor, coexist is located at $665 \pm 5^\circ$ C. and 4.75 ± 0.25 Kb. The compositions of the invariant analcite solid solution, nepheline solid solution, liquid, and vapor were estimated from experiments in the immediate vicinity of the invariant point. The invariant analcite has an anhydrous composition of Ab₅₀Ne₅₀, with between 8 and 9 weight per cent water. The invariant nepheline contains 13 ± 2 weight per cent albite. The invariant liquid has an anhydrous composition of Ab₅₈Ne₄₂, with 15.5 ± 0.5 weight per cent water dissolved in the liquid. The invariant vapor phase contains 5 ± 2 weight per cent dissolved solids with a somewhat higher ratio of nepheline to albite than the invariant liquid. The fact that the invariant liquid contains both more water and albite than the coexisting analcite is of particular importance.

On the basis of the above data the sequence and nature of the five

univariant curves emanating from the invariant point can be given. The reactions along the five univariant curves are: (L)¹ Anl=Ab+Ne+V; (Anl) Ab+Ne+V=L; (V) Anl=Ab+Ne+L; (Ab) Anl+V=Ne+L; and (Ne) Anl+Ab+V=L, given in counterclockwise order starting

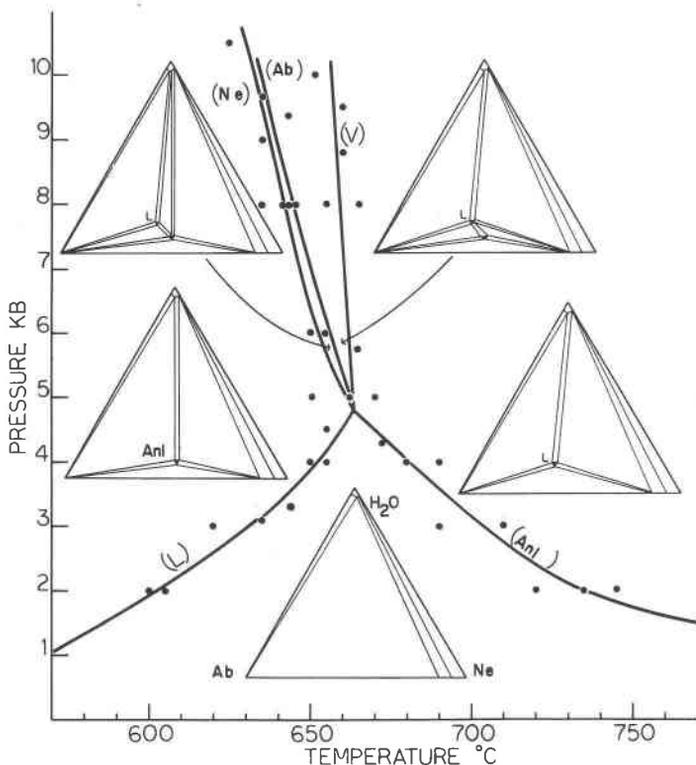


FIG. 2. The invariant point in the system $\text{NaAlSiO}_4\text{-NaAlSi}_3\text{O}_5\text{-H}_2\text{O}$ where analcite solid solution (Anl), nepheline solid solution (Ne), albite (Ab), liquid (L), and a vapor phase (V) coexist, and the related curves of univariant equilibria. The symbols (L), (Anl), (V), (Ab) and (Ne) refer to the reactions along the five univariant curves as discussed in the text. The isobaric-isothermal triangles Ab-Ne- H_2O illustrate the stable phase assemblages in the immediate vicinity of the invariant point and between the curves of univariant equilibria. The solid circles are data points given in Table 3.

from the low pressure decomposition curve for analcite. Thus, three curves extend to higher pressure than the invariant point; (V), (Ab), and (Ne), and two curves extend to lower pressures than the invariant point; (Anl) and (L). Experimentally the curves (L), (Anl), (V), and (Ne) could

¹ Parentheses indicate phase that is absent from the reaction.

TABLE 3. DETERMINATIONS OF THE UNIVARIANT CURVES IN THE SYSTEM NaAlSiO₄-NaAlSi₃O₈-H₂O

Gl=glass Ne=Nepheline Qz=quartz V=vapor Anl=analcite
 tr=trace amounts
 Compositions are in weight %.

Run No.	Composition			Kb Press	°C. Temp.	Days Time	Results
	Anhydrous	H ₂ O	Condition				
38	Ab ₃₈ Ne ₆₂	10	Ab+Ne	8	635	3	Anl+Ne+V?
54	Ab ₄₆ Ne ₅₄	40	Ab+Ne	6	650	4	Anl+V+Ab
55	Ab ₅₃ Ne ₄₇	11.9	Gl	6	650	4	Anl+V
75	Ab ₅₃ Ne ₄₇	14.3	Gl	8	642	2	Anl+V
76	Ab ₅₃ Ne ₄₇	8-9	Anl	8	665	6	Gl+Ne+Ab
91	Ab ₅₃ Ne ₄₇	10.2	Gl	8	645	2	Gl+Ne+V
94	Ab ₃₈ Ne ₆₂	11.7	Ab+Ne	8	645	2	Anl+Ne+Ab
114	Ab ₅₃ Ne ₄₇	23	Ab+Ne	8	665	1	Gl+Ab+V
115	Ab ₅₃ Ne ₄₇	6.1	Gl	8	655	1	Anl+Ab+Ne
122	Ab ₅₃ Ne ₄₇	10.5	Gl	8.8	660	1	Gl+Ne+Ab
154	Ab ₄₆ Ne ₅₄	8-9	Anl	4	655	2	Ab+Ne+V
162	Ab ₅₉ Ne ₄₁	6.3	Ab+Ne	4	680	3	Ab+Ne+V
168	Ab ₅₃ Ne ₄₇	17.8	Gl	5	650	2	Anl+V
188	Ab ₃₈ Ne ₆₂	18	Gl	3.3	645	3	Ne+Ab+V
217	Ab ₅₉ Ne ₄₁	19.2	Ab+Ne	4.3	673	4	Ab+Ne+V
222	Ab ₄₆ Ne ₅₄	20.1	Gl	4	650	1.5	Anl+Ne+V
223	Ab ₅₃ Ne ₄₇	13.4	Gl	4	650	1.5	Anl+V
225	Ab ₅₉ Ne ₄₁	13.1	Gl	4	650	1.5	Anl+Ab+V
227	Ab ₄₆ Ne ₅₄	18.3	Gl	2	605	2	Ne+Ab+V
239	Ab ₄₆ Ne ₅₄	14	Ab+Ne	3	710	2	Gl+Ne+V
241	Ab ₅₉ Ne ₄₁	14.6	Ab+Ne	3	710	2	Gl+Ab+Ne+V
245	Ab ₄₆ Ne ₅₄	19.5	Ab+Ne	2	720	3	Ab+Ne+V
255	Ab ₅₃ Ne ₄₇	15.5	Gl	2	600	2	Ab+Ne+Anl+V
266	Ab ₅₉ Ne ₄₁	16.5	Gl	3	620	2	Anl+V
281	Ab ₄₆ Ne ₅₄	11.6	Gl	3.1	635	4	Ab+Ne+V
284	Ab ₅₃ Ne ₄₇	15.9	Gl	3.1	635	4	Anl+Ab+Ne+V
297	Ab ₄₆ Ne ₅₄	14.6	Gl	4.5	655	2	Anl+Ne+V
299	Ab ₅₃ Ne ₄₇	12.8	Gl	4.5	655	2	Anl+V
311	Ab ₄₆ Ne ₅₄	14.7	Ab+Ne	2	735	2	Ab+Ne+V
312	Ab ₅₅ Ne ₄₅	14.9	Gl	2	735	2	Gl+Ab+V
313	Ab ₅₅ Ne ₄₅	10	Gl	5	663	2	Anl+Ab
314	Ab ₅₅ Ne ₄₅	20	Gl	5	663	2	Gl+Ab+V
320a	Ab ₇₂ Ne ₂₈	15	Gl	1	830	2	Ab+Ne+V
325	Ab ₄₆ Ne ₅₄	14.1	Ab+Ne	9.4	645	1	Gl+Anl+Ne
328	Ab ₅₉ Ne ₄₁	20	Gl	5	662	2	Gl+V
330	Ab ₅₃ Ne ₄₇	20.2	Gl	5	662	2	Anl+Ne+V
331	Ab ₅₃ Ne ₄₇	11.5	Gl	5	662	2	Ab+Ne+Anl
332	Ab ₄₆ Ne ₅₄	20.4	Gl	5	662	2	Gl+Ne+V
334	Ab ₄₆ Ne ₅₄	14.2	Ab+Ne	10	650	1	Gl+Ne+V
337	Ab ₅₉ Ne ₄₁	10.3	Gl	10	650	1	Anl+Ab
338	Ab ₅₉ Ne ₄₁	14.5	Ab+Ne	10	650	1	Gl+Ab+V
345	Ab ₄₆ Ne ₅₄	12.2	Ab+Ne	5	670	7	Gl+Ne+Ab+V
373	Ab ₅₃ Ne ₄₇	16.4	Gl	6	655	3	Anl+Gl
374	Ab ₄₆ Ne ₅₄	14.0	Gl	6	655	3	Anl+Ne+V
377	Ab ₅₃ Ne ₄₇	14.0	Gl	5.5	665	3	Gl+Ne+Ab
378	Ab ₄₆ Ne ₅₄	14.4	Ab+Ne	5.5	665	3	Gl+Ne+V
397	Ab ₅₇ Ne ₄₃	16.3	Gl	5	640	4	Ne+trAnl+V
398	Ab ₄ Ne ₉₆	18.8	Gl	5	640	4	Ne+V
419	Ab ₅₃ Ne ₄₇	10.6	Gl	9.5	660	3	Gl+Ab+Ne
445	Ab ₅₉ Ne ₄₁	17.6	Gl	9.7	635	1	Gl+Ab+V

(Continued on following page)

TABLE 3—(Continued)

Run No.	Composition			Kb Press	°C. Temp.	Days Time	Results
	Anhydrous	H ₂ O	Condition				
447	Ab ₅₃ Ne ₄₇	15.1	Gl	9.7	635	1	Gl+Anl+V
448	Ab ₄₆ Ne ₅₄	12.5	Gl	9.7	635	1	Anl+Ne+V
457	Ab ₄₆ Ne ₅₄	14.6	Gl	9	635	1	Anl+Ne+V
459	Ab ₅₃ Ne ₄₇	16.9	Gl	9	635	1	Anl+V+trAb
463	Ab ₂₀ Ne ₈₀	5.8	Gl	5	640	2	Ne+Anl+V
464	Ab ₁₃ Ne ₈₇	4.4	Gl	5	640	2	Ne+V
485	Ab ₁₆ Ne ₈₄	17	Gl	10.5	625	2	Anl+V
486	Ab ₅₃ Ne ₄₇	17	Gl	10.5	625	2	Anl+V+trAb
487	Ab ₄₀ Ne ₆₀	20	Gl	8	642	3	Anl+Ne+V
489	Ab ₅₃ Ne ₄₇	14.4	Gl	8	642	3	Anl+Gl
490	Ab ₅₃ Ne ₄₇	19	Gl	8	642	3	Anl+Gl+V
500	Ab ₆₅ Ne ₃₅	12.6	Gl	4	690	4.5	Gl+Ab+V
611	Ab ₉₁ Ne ₉	14.6	Gl	5	340	30	Anl+V
612	Ab ₈₈ Ne ₁₂	13.6	Gl	5	340	30	Anl+V
615	Ab ₂₆ Ne ₇₄	16.8	Gl	5	340	30	Anl+V
693-4	Ab ₁₀₀	17.3	Gl	5	300	6	Anl+Ab+Qz+V
712	Ab ₁₀₀	28.4	Ab	5	250	12	Ab+Anl+V
713	Ab ₁₀₀	32.5	Gl	5	250	12	Anl+V

be located quite precisely. The curve (Ab) lies relatively close to the (Ne) curve and could not be located precisely. It is possible, though not experimentally determinable, that a singular point exists on the (Ab) curve where the univariant reaction changes from the $Anl+V=Ne+L$ to $Anl+Ne+V=L$. This feature will be discussed in more detail in the following section.

THE COMPOSITION OF THE UNIVARIANT LIQUIDS

The composition of the first liquid that appeared with increasing temperature in the system was located at a series of pressures; the data are given in Fig. 3a and Table 4. Figure 3a also shows the composition of the eutectic liquid at atmospheric pressure (Greig and Barth, 1938). Edgar (1964) determined the anhydrous composition of the liquid at 1 Kb in the presence of a vapor phase, but did not determine the water content of the liquid. His point is located along the line marked E-E' in Fig. 3a, which does not fit the experimental data of this study (Edgar's temperature is however in good agreement).

A polythermal-polybaric diagram illustrating the changing composition of the univariant liquids along the pertinent P-T curves can be constructed on the basis of the data given in Fig. 3a. This diagram is presented as Fig. 3b where the curves (Anl) and (Ne) represent the changing composition of the univariant liquids as a function of P and T along the P-T univariant curves (Anl) and (Ne), and are taken from Fig. 3a. The (Anl) liquids (Fig. 3b) occur at temperatures above and pressures below

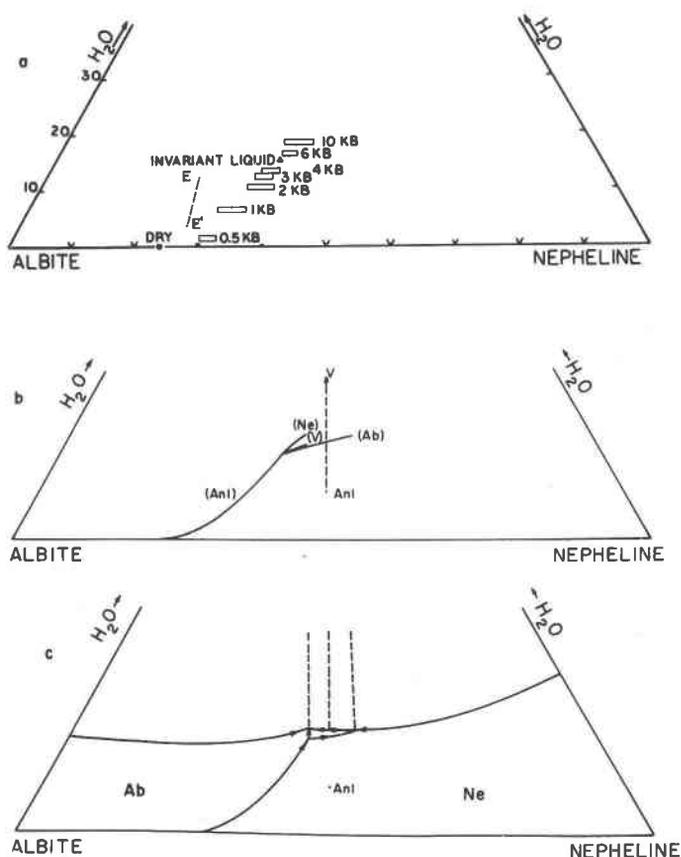


FIG. 3. (a) The composition of the univariant liquid at the beginning of melting. Experimental data are given by rectangles, showing the uncertainty. Edgar's (1964) value at 1 Kb is on the line E-E'.

(b) Polythermal-polybaric composition diagram for the univariant and invariant liquids. The (Ab) and (V) curves are schematic.

(c) Isobaric-polythermal diagram for the probable phase relations at 10 Kb. Decreasing temperature along the boundary curves indicated by arrows. Schematic only.

the invariant point along the univariant P-T curve (Anl). The (Ne) liquids occur at temperatures below and pressures above the invariant point along the univariant P-T curve (Ne). The data given in Fig. 3a and the curve (Ne) in Fig. 3b indicate that the univariant liquid coexisting with Ab-Anl-V along the (Ne) P-T curve becomes increasingly rich in both water and nepheline with increasing pressure. At 10 Kb this liquid lies only slightly towards Ab from the Anl-V join, but does not cross it. The univariant liquid along the (Ab) P-T curve, coexisting with Ne-Anl-V, will contain more nepheline than the (Ne) liquid at a given

TABLE 4. DETERMINATION OF THE COMPOSITION OF THE FIRST LIQUID AT PRESSURES FROM 0.5-10 Kb

Run No.	Composition			Kb Press	°C. Time	Days Time	Results
	Anhydrous	H ₂ O	Condition				
663	Ab ₇₂ Ne ₂₈	5.1	Gl	0.5	925	1.5	Gl+Ab+V
662	Ab ₆₅ Ne ₃₅	5.8	Gl	0.5	925	1.5	Gl+Ne+V
640	Ab ₇₂ Ne ₂₈	2.6	Gl	0.4	950	1	Gl+V
404	Ab ₇₂ Ne ₂₈	4.2	Gl	1	845	5	Gl+Ab+Ne
405	Ab ₇₂ Ne ₂₈	6	Gl	1	845	5	Gl+Ab+trNe
406	Ab ₇₂ Ne ₂₈	7	Gl	1	845	5	Gl+Ab+V
549	Ab ₆₅ Ne ₃₅	6	Gl	1	850	9	Gl
550	Ab ₆₅ Ne ₃₅	7	Gl	1	850	9	Gl+trV
551	Ab ₆₅ Ne ₃₅	4	Gl	1	850	9	Gl+Ab+Ne
552	Ab ₅₉ Ne ₄₁	3.7	Gl	1	850	9	Gl+Ab+Ne
553	Ab ₅₉ Ne ₄₁	7	Gl	1	850	9	Gl+Ne
409	Ab ₇₂ Ne ₂₈	8	Gl	2	765	2	Gl+Ab
414	Ab ₆₅ Ne ₃₅	9.5	Gl	2	745	4	Gl+Ab+Ne
433	Ab ₇₂ Ne ₂₈	9.8	Gl	2	760	6	Gl+Ab+V
654	Ab ₅₉ Ne ₄₁	9.8	Gl	2	750	3	Gl
655	Ab ₅₉ Ne ₄₁	11.7	Gl	2	750	3	Gl+V
657	Ab ₆₂ Ne ₃₈	9.5	Gl	2	750	3	Gl+Ne+trV
658	Ab ₆₅ Ne ₃₅	10.1	Gl	2	750	3	Gl+trV+trAb
193	Ab ₅₉ Ne ₄₁	8-9	Anl	3	720	2	Gl+Ne+Ab
416	Ab ₆₅ Ne ₃₅	8.1	Gl	3	710	3	Gl+Ab+Ne
417	Ab ₆₅ Ne ₃₅	10.6	Gl	3	710	3	Gl+Ab
418	Ab ₆₅ Ne ₃₅	11.5	Gl	3	710	3	Gl+Ab+V
450	Ab ₅₉ Ne ₄₁	9.1	Gl	3	720	3	Gl+Ab+Ne
451	Ab ₅₉ Ne ₄₁	12	Gl	3	720	3	Gl+V
429	Ab ₆₅ Ne ₃₅	10.3	Gl	4	690	2	Gl+Ab+Ne
431	Ab ₅₉ Ne ₄₁	12.7	Gl	4	690	2	Gl+trAb+V?
500	Ab ₆₅ Ne ₃₅	12.6	Gl	4	690	4.5	Gl+Ab+V
501	Ab ₅₉ Ne ₄₁	9.4	Gl	4	690	4.5	Gl+Ab+Ne
502	Ab ₅₉ Ne ₄₁	14	Gl	4	690	4.5	Gl+V+trAb
504	Ab ₅₂ Ne ₄₈	11	Gl	4	690	4.5	Gl+Ne+V
380	Ab ₅₉ Ne ₄₁	13.2	Gl	5	670	2	Gl+Ne+Ab
382	Ab ₆₅ Ne ₃₅	15.2	Gl	5	670	2	Gl+Ab+V
383	Ab ₅₉ Ne ₄₁	14.5	Gl	5	670	2	Gl+trAb
384	Ab ₅₉ Ne ₄₁	17.7	Gl	5	670	2	Gl+trV
385	Ab ₆₃ Ne ₃₇	12.7	Gl	5	670	2	Gl+Ne+Ab
386	Ab ₆₃ Ne ₃₇	15.1	Gl	5	670	2	Gl+Ne+V
616	Ab ₅₉ Ne ₄₁	14.9	Gl	6	680	3	Gl+Ab
617	Ab ₅₉ Ne ₄₁	15.9	Gl	6	680	3	Gl+V
618	Ab ₆₅ Ne ₃₅	14.3	Gl	6	680	3	Gl+Ab
620	Ab ₆₂ Ne ₃₈	16.9	Gl	6	680	3	Gl+Ne+V
539	Ab ₅₉ Ne ₄₁	19.9	Gl	10	640	2	Gl+Ab+V
541	Ab ₅₂ Ne ₄₈	19.4	Gl	10	640	2	Gl+trV
591	Ab ₆₂ Ne ₃₈	18.3	Gl	10	655	5	Gl
593	Ab ₄₆ Ne ₅₄	17.6	Gl	10	655	5	Gl+Ne+V

pressure. Thus, it seems quite possible that the (Ab) univariant liquid will cross the Anl-V join at a unique pressure and temperature along the (Ab) univariant P-T curve. If this occurs, then the singular point mentioned in the previous section is developed. At this pressure and temperature the four-phase univariant reaction changes from Anl+V=Ne+L (lower pressure) to Anl+Ne+V=L (higher pressure). Thus, the univariant liquid coexisting with Anl-Ne-V along the (Ab) P-T curve lies

on the Anl-V join at the pressure and temperature of the singular point (Fig. 3b). At pressures below the singular point on the (Ab) P-T curve the univariant liquid lies outside the triangle formed by Anl-Ne-V toward Ab. At pressures above the singular point the univariant liquid on the (Ab) P-T curve lies within the triangle Anl-Ne-V. At this unique pressure and temperature another P-T curve is generated, extending to higher pressures. This curve is governed by the degenerate (in a ternary system) reaction $\text{Anl} + \text{V} = \text{L}$. The univariant liquid coexisting with Ne-Ab-Anl also changes composition with increasing pressure along the (V) univariant P-T curve. This change is indicated in Fig. 3b by the line labeled (V). As in the case of the (Ab) liquid, this curve (V) is located only approximately.

In the previous section we have discussed the change in composition of the various univariant liquids along particular P-T curves of univariant equilibria. It should be noted that analcite, nepheline, albite, and vapor are expected to change composition in an analogous manner along the P-T curves of univariant equilibria. Thus four curves radiate from each of the four compositions representing the invariant Anl, Ne, Ab and V. Each of these sixteen curves then represents, polythermally and polybarically, the changing composition of a univariant phase along a particular P-T curve of univariant equilibria. These curves are not presented in this paper because of the lack of information on their compositions.

An isobar at a pressure greater than the invariant point would intersect the (Ne), (V) and (Ab) curves mapping out a triangular area. The three curves bounding this triangular region represent the composition of the bivariant liquids which coexist with (1) Anl+V, (2) Anl+Ab and (3) Anl+Ne. Thus these three curves bound the primary field of Anl which is the triangular region. The three curves then are isobaric boundary curves. In order to present a more complete isobaric diagram we also need the boundary curves where liquids coexist with (1) Ab+V, (2) Ne+V, and (3) Ab+Ne. Figure 3c illustrates the most probable configuration of the isobaric diagram at 10 Kb, based on the previous discussion. We see that the join Anl-V represents a temperature maximum for water saturated liquids. Again this is assuming the existence of the singular point at some pressure between 7 and 10 Kb. It is apparent that the (Ab) P-T curve can cross the (Ne) P-T curve if the singular point exists. If this intersection of the two curves does take place, it should be noted that the curves will intersect in projection only since the two liquids along the curves are not of the same composition, nor do they coexist. If the singular point does not exist and the two curves meet, an invariant point of essentially the same type as found at $665 \pm 5^\circ \text{C}$. and 4.7 Kb will occur which seems somewhat unlikely.

Vapor phase. The vapor phase was studied to see if its composition re-

mains in the three component system $\text{NaAlSi}_3\text{O}-\text{NaAlSiO}_4-\text{H}_2\text{O}$. The vapor phase at 6 Kb and $675^\circ\text{C}.$ - $680^\circ\text{C}.$, a temperature and pressure just above the invariant point, was studied in detail. The principal results are tabulated in Table 5 and shown in Fig. 4.

The SiO_2 -rich portion of the vapor phase crystallizes on the quench partly into big idiomorphic albite crystals that can be distinguished from

TABLE 5. DETERMINATION OF THE VAPOR COMPOSITION AT 6 Kb AND 675 - $680^\circ\text{C}.$

Run No.	Composition			Kb Press	$^\circ\text{C}.$ Temp.	Days Time	Results
	Anhydrous	H_2O	Condition				
529	Ab ₉₁ Ne ₉	71	Gl	6	680	7	Ab+V
530	Ab ₈₆ Ne ₃₈	66	Gl	6	680	7	Gl+Ab+V
531	Ab ₈₂ Ne ₄₈	71	Gl	6	680	7	Gl+V
532	Ab ₅₀ Ne ₆₀	69	Gl	6	680	7	Gl+Ne+V
533	Ab ₄₆ Ne ₅₄	70.3	Gl	6	680	7	Gl+V
535	Ab ₁₃ Ne ₈₇	68.3	Gl	6	680	7	Ne+V+trGl
556	Ab ₅₉ Ne ₄₁	88.5	Gl	6	675	6	Ab+Gl+V
558	Ab ₂₆ Ne ₇₄	90	Gl	6	675	6	Gl+V
559	Ab ₈₅ Ne ₁₅	96.5	Gl	6	675	6	V
560b	Ab ₂₈ Ne ₇₄	95	Gl	6	675	6	V
570	Ab ₈₆ Ne ₁₄	48.7	Gl	6	675	4	Ab+Gl+V
579	Ab ₁₃ Ne ₈₇	48.4	Gl	6	675	4	Ne+V
580	Ab ₈₅ Ne ₁₅	80	Gl	6	675	4	Ab+V
581	Ab ₈₅ Ne ₃₅	79.5	Gl	6	675	4	Gl+Ab+V
582	Ab ₅₉ Ne ₄₁	79.2	Gl	6	675	4	Gl+V+trAb
584	Ab ₄₆ Ne ₅₄	80.5	Gl	6	675	4	Gl
587	Ab ₁₃ Ne ₈₇	80.3	Gl	6	675	4	Ne+V+Gl
600	Ab ₁₃ Ne ₈₇	95.6	Gl	6	680	8	V
601	Ab ₁₃ Ne ₈₇	90.4	Gl	6	680	8	Gl+V+Ne?
604	Ab ₇₂ Ne ₂₈	90.8	Gl	6	680	10	Ab+V
605	Ab ₇₂ Ne ₂₈	92.8	Gl	6	680	10	V+trAb
606	Ab ₅₉ Ne ₄₁	92.3	Gl	6	680	10	V
607	Ab ₈₅ Ne ₁₅	93.1	Gl	6	680	10	Ab+V
622	Ab ₁₀₀	96	Gl	6	680	11	V
625	Ne ₁₀₀	92.6	Gl	6	680	11	Ne+V
626	Ne ₁₀₀	95.5	Gl	6	680	11	V
627	Ab ₁₃ Ne ₈₇	93.3	Gl	6	680	11	Gl+V
666	Ab ₄₈ Ne ₅₄	93.2	Gl	6	680	9	V
667	Ab ₂₃ Ne ₆₇	91	Gl	6	680	9	Gl+V
668	Ab ₂₅ Ne ₇₄	93.7	Gl	6	680	9	V+Gl
705	Ab ₇₂ Ne ₂₈	86.5	Gl	6	675	4	Ab+Gl+V
707	Ab ₅₂ Ne ₄₈	92	Gl	6	675	4	V+Gl

the fine grained aggregates of the equilibrium albite phase. Nepheline does not form on the quench, but large nepheline crystals are encountered just outside the vapor field boundary, above 90% H_2O . The relative amounts of the phases forming from the quenched vapor, glass, albite, corundum and sodium carbonates, varies with the anhydrous starting compositions. The small amounts of paragonite that were now and then detected with the glass and nepheline in Si-poor and H_2O -rich part of the system may be an indication that the system is not strictly ternary over

the studied temperature and pressure range, although it may also represent metastable crystallization. It is, however, over only a very limited part of the system, so that it can be ignored for the purpose at hand.

The two-phase regions albite-vapor, liquid vapor and nepheline-vapor are moved at high H_2O contents toward the sideline $NaAlSiO_4-H_2O$. Generally, therefore, the composition of the vapor is somewhat poorer in SiO_2 than the coexisting phases. The vapor-phase at the nephe-

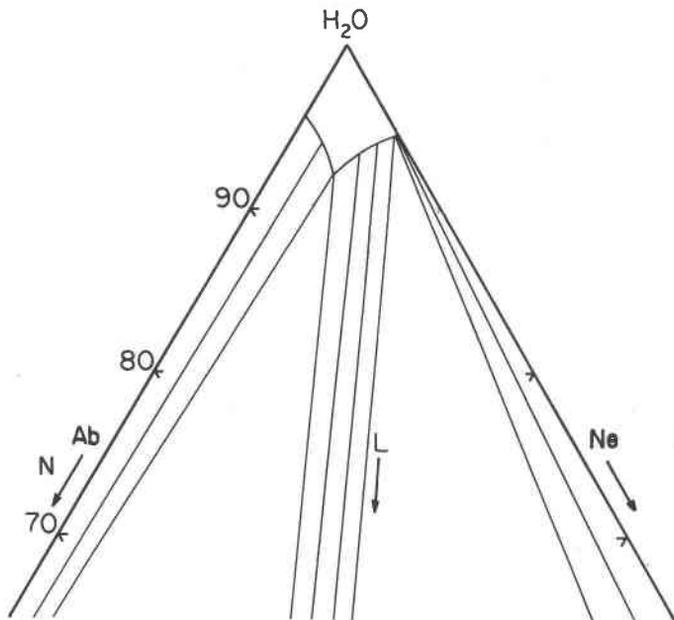


FIG. 4. The composition of the vapor phase at 675° C., 6 Kb. Assemblages encountered are: Vapor, nepheline solid solution+vapor, nepheline solid solution+liquid+vapor, liquid+vapor, albite+liquid+vapor, and albite+vapor.

line side of the system may be enriched in sodium as well, as indicated by the appearance of paragonite.

Stability of analcite solid solutions. Saha (1961) indicated that analcite with the composition $Na_2Al_2Si_{3.25}O_{10.5} \cdot nH_2O$ is the most stable¹ composition and not analcite with the ideal composition $Na_2Al_2Si_4O_{12} \cdot 2H_2O$. Greenwood (1961) found a value of 3.5 for the molecular ratio of SiO_2 to Na_2O in the analcite he synthesized. In the course of this study it was found that the most stable analcite has a composition between $Na_2Al_2Si_{3.25}O_{10.5} \cdot nH_2O$ and $Na_2Al_2Si_{3.5}O_{11} \cdot nH_2O$, somewhat nearer to the latter

¹ We refer to the composition of the analcite solid solution which is stable at the highest temperature for a given pressure as the "most stable" composition.

composition. Expressed in weight per cent albite and nepheline the anhydrous composition is about $Ab_{50}Ne_{50}$. The value $\Delta(2\theta_{An1(639)}-2\theta_{Si(331)})$ for Cu $K\alpha_1$) for the most stable analcite varies between 1.59 and 1.62 degrees 2θ .

Analcite with the stoichiometric composition $Na_2Al_2Si_4O_{12}\cdot 2H_2O$ is stable at 1 Kb up to about $530^\circ C.$ and at 5 and 10 Kb up to about $550^\circ C.$ Above these temperatures analcite reacts to form an analcite richer in nepheline, and albite. The change from an unstable analcite solid solution

TABLE 6. DATA ON THE SYSTEM $NaAlSiO_4$ - $NaAlSi_3O_8$ - $KAlSi_3O_8$ - H_2O

Or = $KAlSi_3O_8$ F = feldspar Ne = nepheline Anl = analcite Gl = glass
V = vapor Tr = trace

Run No.	Composition			Kb Press	°C. Temp.	Days Time	Results
	Anhydrous	H ₂ O	Condition				
722	$Ab_{29}Or_{40}Ne_{31}$	20	Gl	2	670	1	Ne+F+Gl+V
740	$Ab_{29}Or_{40}Ne_{31}$	29.5	Gl	1	720	1	Ne+F+Gl+V
741	$Ab_{29}Or_{40}Ne_{31}$	23.1	Gl	2.8	625	1	Ne+2F+V
746	$Ab_{29}Or_{40}Ne_{31}$	36.7	Ne+F	2	660	2	Ne+F+Gl+V
744	$Ab_{29}Or_{40}Ne_{31}$	25.1	Gl	2	650	2	Ne+F+V
754	$Ab_{29}Or_{40}Ne_{31}$	35.8	Gl	6	612	2	Anl+Ne+F+V
759	$Ab_{29}Or_{40}Ne_{31}$	36.8	Ne+F	6	612	2	Ne+Anl+F+V
761	$Ab_{29}Or_{40}Ne_{31}$	40	Ne+F	3.2	645	2	Ne+F+V+trGl
762	$Ab_{29}Or_{40}Ne_{31}$	35.8	Ne+F	3.25	640	2	Ne+F+V
785	$Ab_{48}Or_{12}Ne_{44}$	14.7	Gl	6.4	630	3	Anl+V
786	$Ab_{29}Or_{40}Ne_{31}$	19	Gl	6.4	630	3	Gl+Anl+Ne+V
792	$Ab_{48}Or_{12}Ne_{44}$	26.2	Gl	1.7	655	2	Ne+F+V
798	$Ab_{48}Or_{12}Ne_{44}$	23.8	Gl	3.25	640	2	Anl+V
799	$Ab_{48}Or_{12}Ne_{44}$	21.7	Gl	7.5	650	2	Gl+V
800	$Ab_{48}Or_{12}Ne_{44}$	30.6	Gl	2.2	635	2	Anl+V
801	$Ab_{48}Or_{12}Ne_{44}$	26.1	Gl	1	610	2	Anl+V+Ne+trF
814	$Ab_{48}Or_{12}Ne_{44}$	29.1	Gl	1	620	2	Ne+F+V
815	$Ab_{48}Or_{12}Ne_{44}$	29.7	Gl	1.5	630	2	Ne+F+V

to a stable analcite solid solution takes place very slowly and in most cases glasses were used to determine the stability of the analcite solid solutions. The reaction products were generally the same whether the starting material was a glass, analcite or a mixture of albite and nepheline. Equilibrium, however, was attained much faster with glass as the starting material.

Addition of potassium to the system albite-nepheline- H_2O . A preliminary investigation of the system $NaAlSi_3O_8$ - $NaAlSiO_4$ - $KAlSi_3O_8$ - H_2O was undertaken to see how the addition of potassium to the system albite-nepheline-water affects the stability of analcite and the minimum melting temperature. Some of the results are tabulated in Table 6. It was found that analcite solid solutions are stable at 1 Kb up to $610^\circ C.$ and at 2 Kb up to $640^\circ C.$ and that the minimum melting temperature was $730^\circ C.$ at

1 Kb, 655° C. at 2 Kb, and 645° C. at 3 Kb. Thus, the melting curve is lowered considerably and the stability of analcite increased compared to the system albite-nepheline-water. The melting curve and the stability field of analcite meet at about 2.3 Kb and 650° C. Thus by adding potassium to the system, analcite and liquid can coexist at much lower pressures and temperatures, than in the system albite-nepheline-water.

Analcite crystallized from the starting material $\text{Ab}_{43}\text{Or}_{13}\text{Ne}_{44}$ in the vapor present region did not have detectable amounts of other crystalline phases. Those that crystallized from somewhat potassium-richer starting materials were accompanied by considerable amounts of nepheline and feldspar. This indicates that small amounts of potassium (about 2 weight per cent K_2O) can be taken up in the analcite solid solutions. It was also found that these small amounts of potassium decrease the cell edge which suggests that the analcite-determination curve should be used with caution for natural analcites. Analcites occurring in igneous rocks contain potassium in amounts from 0.50 to 4.48 weight per cent K_2O .

DISCUSSION AND PETROGENETIC APPLICATIONS

In igneous rocks analcite can occur in several different ways: Analcites from amygdules in basalts crystallized in a late hydrothermal stage and are of secondary origin. Analcite replacing other minerals like leucite (Burgess, 1941), alkali feldspars and plagioclase feldspar (Jahns, 1938; Lonsdale, 1940; Wilkinson, 1962) and nepheline (Tyrrell, 1917) is also of secondary origin. In the cases where analcite is interstitial in teschenites (Tyrrell, 1912), lugarites (Tyrrell, 1917), syenites and syenogabbros (Lonsdale, 1940), tinguaites (Wilkinson, 1963) and basalts (Tomita, 1933; Wilkinson, 1962) it is generally accepted, that the analcite formed in the later stages of crystallization from alkali-rich differentiates. Analcite phenocrysts described from mafic phonolites (Pirsson, 1905; Larsen, 1941), blairmorites (MacKenzie, 1914) and trachybasalts (Lyons, 1944) show no evidence of being pseudomorphs after leucite and are probably crystallized at liquidus temperatures.

The experiments show that analcite can crystallize directly from a melt in the system albite-nepheline- H_2O at pressures above 4.5–5 Kb and can coexist with the liquid over a small temperature range. The addition of potassium lowers this pressure and temperature considerably. Analcite phenocrysts may crystallize in the small temperature range, where analcite coexists with alkali-rich melts. On the further cooling of these alkali- and water-rich melts the groundmass analcite, which accompanies the analcite phenocrysts, could crystallize well within its stability field. Most analcite-bearing igneous rocks contain analcite only in the groundmass. Here the analcite seems to have been formed during the rapid cool-

ing from the alkali-rich mesostasis of basic rocks. The experiments suggest that it is not necessary that a PT-field has to be passed where analcite is metastable, as has been proposed by Wilkinson (1962) for the analcite basalts of Spring Mount in New South Wales. This assumption was based on the preliminary studies of the system albite-nepheline-H₂O, where the invariant point lies at a rather high pressure as demonstrated in this study.

The extent of solid solution in the synthetic analcites decreases with increasing temperature. One should expect, therefore, that the primary analcites in igneous rocks have a very restricted range in composition, in contrast to the analcites in sedimentary rocks. As a matter of fact, the data compiled by Wilkinson and Whetton (1964) and Whetten and Coombs (1965) for compositions of sedimentary analcites show a very wide range in composition. For sedimentary analcites it seems as if the effects of chemical environment are more pronounced than those of temperature with respect to the composition of the analcites. There are not many analyses of analcites occurring as primary constituents in igneous rocks. The few existing analyses (MacKenzie, 1914; Larsen and Buie, 1938; Wilkinson, 1963) give compositions ranging from Na₂Al₂Si₃O₁₀·nH₂O to Na₂Al₂Si₄O₁₂·2H₂O. This agrees fairly well with the experimental data showing that analcite with composition Na₂Al₂Si_{3.4}O_{10.8}·nH₂O is most stable. However, there are still too few analyses of primary analcite to draw definite conclusions. Also the role of other elements such as potassium is not yet well known and must be investigated further.

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