The Composition of Nepheline

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With 2 figures in the text and 5 tables

The crystalline lattice

The crystal structure of nepheline was determined by BUERGER, KLEIN and DONNAY (1954) and refined by HAHN and BUERGER (1955). The dimensions of the hexagonal unit cell change with the composition (SMITH and SAHAMA, 1954; SMITH and TUTTLE, 1957; DONNAY et. al., 1959); most natural nepheline of non-extrusive rocks exhibit values close to: $a_0 = 10.00$ Å, $c_0 = 8.38$ Å.

In this cell there are 32 oxygens in a tetrahedral arrangements around 16 cations of Al and Si. This part of the cell, which can be written $(Si, Al)_{16}O_{32}$, corresponds to the structure of tridymite, and, like tridymite, it inverts into a cristobalitelike structure at elevated temperatures $\simeq 1280°$) socalled carnegierite (BARTH and POSNJAK, 1932). Into the open cavities of the tridymite (and cristobalite) lattice enter the larger cations: Na, K, Ca, thus balancing the valences of the $[(Si, Al)_{16}O_{32}]^{8+}$ framework. There are 8 structural sites for these cations, and 8 valences to saturate. If 8 univalent cations — Na or K — enter into the cell, they will fill all available sites and also satisfy the valences. However, it is an interesting detail of the structure proposed by HAHN and BUERGER, that 2 of the cavities are larger than the others and therefore preferred by K ions, whereas the remaining 6 are smaller and preferred by Na (and Ca) ions. Consequently, the proportion K : (Na + Ca) might be expected to approach 1 : 3. However, in natural nepheline the presence of additional ions modify this statement, v. i.

If bivalent Ca enters into the cavities, just 4 Ca are sufficient to balance the valences, and so the additional 4 cavities remain open.

An excess of SiO_2 may also enter into the nepheline structure. It can be looked upon as a molecule of $Si_{16}O_{32}$. In this case there are no

extra valences to balance — all 8 structural sites in the middle of the cavities will, therefore, remain open.

The constituent mineral molecules

Thus we may look upon the nepheline structure cell as accommodating 4 mineral molecules plus holes ($\square$)[1].

24 Ne	(Nepheline)	Na_4	Na_4	Al_8	Si_8	O_{32}
24 Ks	(Kalsilite)	K_4	K_4	Al_8	Si_8	O_{32}
24 An' = 20 An + 4 $\square$		$\square_4$	Ca_4	Al_8	Si_8	O_{32}
24 Q' = 16 Q + 8 $\square$		$\square_4$	$\square_4$	Si_8	Si_8	O_{32}

In the following, also the holes — which are potential sites for cations — will be reckoned as cations and included in the equivalent molecular percentages. It is best, however, to separate the holes into a group for themselves. We thereby arrive at the following 5 components of nepheline, Ne, Ks, An, Q, $\square$. (Nepheline-Kalsilite-Anorthite-Quartz-Holes). It should be emphasized, however, that these 5 components represent only a 4-component system, for the amounts of holes cannot vary independently, but is determined by the relation: $\square = {}^1/_5$ An $+ {}^1/_2$ Q.

The following additional molecules might be considered as possible components of nepheline.

1. Calcium aluminate, which can be written $Ca_4Ca_4Al_8Al_8O_{32}$. It would explain the extra Al_2O_3 which is found in some analyses. However, also water, in form of hydroxonium ions may bind Al_2O_3 (v. i.), which makes it unnecessary to introduce calcium aluminate.

2. Sodium aluminate, $Na_8Na_8Al_8Al_8O_{32}$, exhibits at high temperature the cristobalite structure (the extra Na ions go into still another set of crystallographically possible sites in the unit cell, Barth 1935) and forms a continuous series of mixed crystals with carnegieite (Schairer 1947). In natural nepheline, however, this substitution does not seem to take place.

3. Hydroxonium nepheline. In most nepheline analyses there are rather large amounts of H_2O. Very little attention has been paid to this fact by recent investi-

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[1] In this paper are used the equivalent molecular units as first introduced by P. Niggli (1936), or the one-cation unit as they were later called by Eskola (1954). Equivalent formula units are defined as those which contain the same number of cations, and it is simplest to reduce the several equivalent units to a sum of the cations = 1. Then the sum of all cations in the formula represents the number of equivalent units. Consequently, $Na_2O \cdot Al_2O_3 \cdot 2SiO_2 = 6$ Ne (because the formula thus written contains 6 cations); $NaAlSiO_4 = 3$ Ne, and $SiO_2 = 1$ Q (because the formulas contain 3 and 1 cation respectively).

gators. But the older mineralogists were aware of it. In HINTZE's Handbuch der Mineralogie (1897) "Hydronephelit" is listed as a valid species — uniaxial positive with composition $HNa_2Al_3Si_3O_{12} \cdot 3H_2O$; BOWEN (1917), the first to express rather correctly the composition of nepheline in terms of constituent molecules, simply mentions "that H_2O seems to replace the alkaline oxides to a moderate extent".

DUNHAM (1933) found hydronephelite on the Hawaiian Islands, and NEUMANN (1949) discussing the known data suggested that hydronephelite-ranite are not zeolites but constitute a mineral group different from that of the zeolites.

The nepheline analyses of this paper are best recalculated by assuming that hydroxonium replaces potassium. The replacement of (SiO_4) by (H_4O_4) as found in e. g. hydrogarnets does not fit the analytical data. There are always only small amounts of hydroxonium in nepheline; a theoretical end member could be written:
$Hn = (H_3O)_4 (H_3O)_4 Al_8 Al_8 [O_{24}/OH_8]$.

It was mentioned that the unit cell has 2 sites for K and 6 sites for Na, but that in natural nephelines this proportion is influenced by the other ions present. It would seem that the 2 large cavities per unit cell are willing to accept potassium ions or no ions at all[2]. The function of these two sites is therefore not so much to accept K as it is to exclude Na and Ca. What happens is that the 6 smaller cavities easily fill up with Na and Ca, whereas the amount of K permitted into the larger cavities is limited to the amount balancing the valences. Presence of Ca, or of an excess of SiO_2, makes some of the large cavities remain open, thus reducing the theoretical 1 : 3 proportion of K : (Na + Ca). HAHN and BUERGER find that in most natural nepheline Ca is present in amounts approximately corresponding to 0.4 atoms per unit cell, and they give the ideal (or average) formula of natural nepheline as $K_{1.2}Ca_{0.4}Na_{5.6} Al_{7.6}Si_{8.4}O_{32}$. If we also want to indicate the "holes", the formula becomes

$$Na_{5.6}K_{1.2}Ca_{0.4}\square_{0.8} [Al_{7.6}Si_{8.4}O_{32}].$$
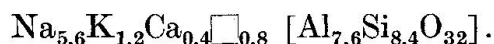
It is therefore the sum $(K + \square)$ which occupy 2 sites in the unit cell[3], while (Na + Ca) occupy 6 sites.

Expressed on a percentage basis in terms of mineral molecules, we must consider that the symbol Ks as used in this paper includes $1/3$ K, consequently,

$$Ks + 3\square \simeq 25\% \qquad (1)$$

The remaining part of the structure is $Ne + An + Q - 2\square = 75\%$. As previously explained the number of holes created in the structure has

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[2] It is worthy of note that Ba or Sr of similar radii are not accepted, but they do enter into the associated feldspars. See analyses 3, 4 and 5'.

[3] Also hydroxonium ions will occupy the large sites. If one wants to include water in the formula, it would be the sum $K + \square + (H_3O)$ which should make 2.

Table 1. *The nepheline formula of* HAHN *and* BUERGER *(H and B) and of*
MOROZEWICS *(M) recalculated into mineral molecules*

| | Number of equivalent molecules in unit cell | | Molecular % | |
	H and B	M	H and B	M
Ne	16.8	18.3	70.0	76.2
Ks	3.6	4.5	15.0	18.8
An	2.0	—	8.3	—
Q	0.8	0.8	3.3	3.3
□	0.8	0.4	3.3	1.7
Sum	24.0 cations	24.0 cations	99.9 %	100.0 %

the following relation: $\square = {}^1/_5\,\mathrm{An} + {}^1/_2\,\mathrm{Q}$; by substitution one derives

$$\mathrm{Ne} + {}^3/_5\,\mathrm{An} \cong 75\% \tag{2}$$

which is complimentary to equation (1).

The chemical composition

The chemical equations (1) and (2) restrict the chemical composition of nepheline from gneisses, pegmatites, and most plutonic rocks. They give a crystal-structural explanation of the nepheline "formula" proposed by MOROZEWICS, who for a half century staunchly defended his thesis of a constant nepheline composition against a united opposition from other leading mineralogists. Now the tide has turned. MIYASHIRO (1951) investigated the chemical composition of nephelines and found that all nepheline from non-effusive rocks exhibited a surprisingly narrow range of chemical variations.

TUTTLE and SMITH (1958) investigated the phase relations in the Ne-Ks system, and found the solvus at lower temperatures to follow the composition $Na_3KAl_4Si_4O_{16}$. They also indicated, although no experimental proof was at hand, that there may be a miscibility gap between this nepheline ($Na_3KAl_4Si_4O_{16}$) and the end member ($NaAlSiO_4$). This is as much as saying that $Na_3KAl_4Si_4O_{16}$ is a compound at low temperatures. But even at low temperature the compound is able to take some SiO_2 and some CaO (as $CaAl_2Si_2O_8$) in solid solution, thereby the ratio K : Na is somewhat reduced, as previously explained.

An inspection of Tables 2 and 3 and of Figures 1 and 2 supports this assumption: Alle nephelines show a remarkable constant composition

Table 2. *Chemical analyses of nephelines and feldspars from the Seiland-Stjernö province, N. Norway*

	Nephelines, weight percent				Feldspar, weight percent			
	1	2	3	4	1'	2'	3'	5'
SiO_2	42.85	43.12	43.34	43.66	64.90	65.62	66.42	66.25
Al_2O_3	33.36	34.28	32.81	34.05	18.98	21.60	20.41	19.75
Fe_2O_3	0.10	0.11	0.39	0.12	0.05	0.06	0.28	0.29
CaO	2.31	0.36	0.56	0.28	0.13	1.64	1.03	0.60
Na_2O	15.00	17.03	16.26	15.60	3.84	10.03	10.20	8.10
K_2O	5.81	5.04	4.63	5.96	11.16	1.19	1.04	4.44
H_2O^+	—	0.11	1.06	0.52	0.10	0.02	0.23	0.09
	100.15	100.13	99.92	100.29	99.76	100.21	99.83	100.11

1 Includes $CO_2 = 0.72\%$; 2 includes $H_2O^- = 0.08\%$; 3 includes $MnO = 0.02$, $MgO = 0.07$, $Cl = 0.11$, $SO_3 = 0.04$, $CO_2 = 47$, $H_2O^- = 0.21$, not present are: Ti, Ba, Sn, P; 4 includes $MgO = 0.10$. Feldspar analyses: 1' Includes $BaO = 0.66$; 2' includes $TiO_2 = 0.02$, $MgO = 0.01$, $P_2O_5 = 0.02$; 3' includes $BaO = 0.11$, $SrO = 0.11$; 5' includes $BaO = 0.39$, $SrO = 0.067$, $MgO = 0.09$, $H_2O = 0.04$, $CO_2 = $ nil.

Additional analyses from Bekkarfjordnes (4)

		Nephelines						Feldspar						
		4a	4b	4c	4d	4e	4f		4b'	4c'	4d'	4g'	4h'	4i'
Weight	% Na_2O	15.20	15.52	16.60	15.40	15.40	15.20		8.68	8.89	10.18	7.92	10.10	8.46
	% K_2O	5.48	5.50	5.65	5.60	5.98	5.10		4.24	3.92	1.40	4.66	1.60	3.84
	% CaO	—	—	—	—	—	—		0.60	0.88	—	—	—	—
Proportions	Ne	81	81	82	81	79	82	Ab	74	77	91	72	91	77
	Ks	19	19	18	19	21	18	Or	23	19	9	28	9	23
								An	3	4	—	—	—	—

Description of specimens from Seiland and Stjernö

The islands of Seiland and Stjernö are located southwest of North Cape, Northern Norway, at about 90° 30' N, 12° E.

1 and 1': Nepheline and feldspar, respectively, from nepheline syenite, Lille-bukt, Stjernö. The rock has been described by HEIER (1961), the chief minerals are orthoclase perthite and nepheline with smaller amounts of biotite, calcite, sphene, apatite, magnetite.

2 and 2': Nepheline and feldspar, respectively, from nepheline syenite-pegmatite

dike Kufjord, Seiland. The chief minerals of the dike are albite antiperthite and nepheline with additional biotite (about 8%) and magnetite.

3 and 3': Nepheline and feldspar, respectively, from ringite pegmatite, Skarve-bukt, Seiland. The pegmatite has been described by BARTH (1927), and consists of huge crystals of albite antiperthite, nepheline, calcite, green apatite, brown apatite, biotite, and small amounts of sphene and magnetite. The nepheline is in places penetrated by an irregular fine net work of sodalite.

4 and 4': Nepheline and feldspar, respectively, from nepheline syenite pegma-tite, Bekkarfjordnes, Seiland. The rock consists of antiperthitic alkali feldspar and nepheline with smaller amounts of biotite, magnetite, calcite and nepheline.

5': Feldspar from syenite pegmatite (antifenite, BARTH, 1927), Hakkstabben, Seiland. In this rock antiperthitic alkali feldspar is strongly dominant; there are small amounts of biotite and magnetite.

Table 3. *Cations to 32 (O+OH) in nepheline* (Fe is included in Al)

	1	2	3		4
Si	834	826	838[2])	846[2])	833
Al	767	774	747	754	767
Ca	29[1])	7	12	—	6
Na	567	633	609	596	577
K	144	123	115	118	144
H_2O	—	7	67	68	33

Constituent molecules of the nephelines

	Cell	Mol%	Cell	Mol%	Cell	Mol%	Cell	Mol%
Ne	17.0	70.8	19.0	79.1	17.9	74.5	17.3	72.2
Ks	4.3	18.0	3.7	15.4	3.5	14.6	4.3	18.0
An	1.45	6.0	0.35	1.45	—	—	0.3	1.2
Q	0.65	2.7	0.56	2.3	1.3	5.4	1.0	4.2
Hn	—	—	0.1	0.4	0.6	2.5	0.5	2.1
	0.60	2.5	0.3	1.4	0.7	2.9	0.6	2.3
	24.00	100.0	24.01	100.05	24.0	99.9	24.0	100.0

Composition of associated feldspar

	Or	Ab	An	Or	Ab	An	Or	Ab	An	Or	Ab	An
	65.2	34.2	0.6	6.4	85.9	7.7	6.0	89.1	4.9	23	73	4

[1]) After subtraction of CO_2.

[2]) The first column gives the proportions directly from the analysis, the second column is adjusted for hematite 0.3%, biotite 0.3%, sodalite 1.8%, calcite 1.0%.

in spite of the great variation in the ratio K : Na of the several rocks in which they occur: nephelines of essentially the same composition are stable in contact both with practically pure albite and with potassic microperthite. This is precisely the property to be expected of a compound: The activity of Na in nepheline (of almost constant composition) will change from 1 (as defined by the activity of Na in albite) to near zero (defined by orthoclase perthite, or by almost pure kalsilite as shown in TUTTLE and SMITH's experiments).

But at higher temperatures $Na_3KAl_4Si_4O_{16}$ is no longer a compound. This was clearly shown by TUTTLE and SMITH. Above ca. 500° C the solid solution series becomes appreciable and at 800—900° it extends far toward the $KAlSiO_4$ end member. This is amply supported by the
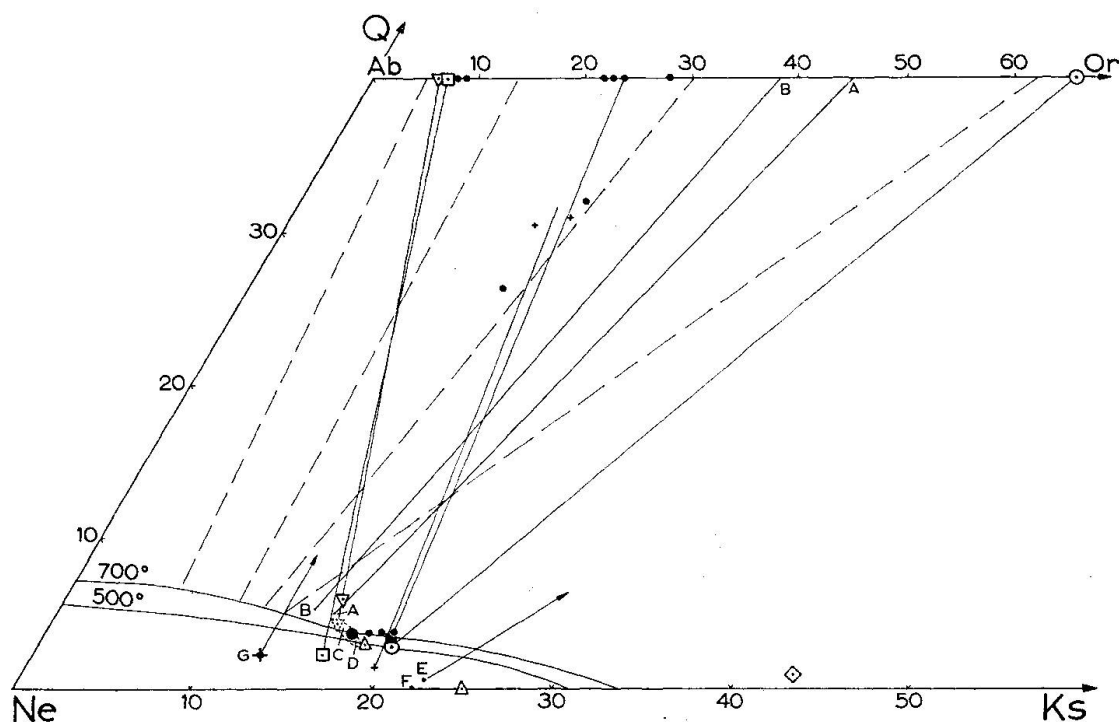

Fig. 1. Projection points of the chemical composition of nephelines and feldspars in the ternary diagram Quartz-Nepheline-Kalsilite.
● Hahn-Buerger formula, see Table 1. ■ Morozewics' formula, see Table 1. O Stjernö, ☐ Kufjord, ▽ Skarvebukt, • Bekkarfjordnes, corresponding respectively to nos. 1, 2, 3, 4 of Tables 2 and 3.
A, B, C, D, E, F, G as in table 4. △ is H, and + is I of Table 4. ⟨·⟩ is nepheline from lava at Mount Nyiragongo (SAHAMA 1953a). Coexisting nephelines and feldspars are joined by tie lines. The experimental data of Hamilton and MacKenzie are indicated as follows: The limits of solid solutions are drawn for 500° and 700°, and the composition of coexisting pairs of nepheline-feldspars at 700° are joined by stippled lines.

Table 4. *Additional nepheline analyses used in this paper*

	A	B	C	D	E	F	G	H	I
Ne	75.4	76.6	75.6	75.5	76.0	75	79.1	74.3	75.6
Ks	14.4	13.2	15.1	16.5	22.5	21	14.5	17.1	20.2
An	2.5	2.1	2.4	3.0	0.6	4	2.7	3.1	1.0
Q	4.8	5.1	4.0	3.0	0.5	—	2.1	3.0	2.1
Hn	—	—	0.4	—	—	—	—	0.5	—
□	2.9	3.0	2.6	2.0	0.3	1	1.6	2.0	1.2

A and B are from Bratholmen and Laagendal, respectively, South Norway;
C and D are from Monmouth, Ontario, corresponding to Nos. 65270 and 82464;
TILLEY and GITTINS, 1961. E and F are from Loyne, Sutherland, and Pilandsberg,
Transvaal, respectively; TILLEY 1956. G is from Mogok, Burma, TILLEY 1954.
H is from York River, Ontario, ZIES and CHAYES 1960. I is from Blue Mountain,
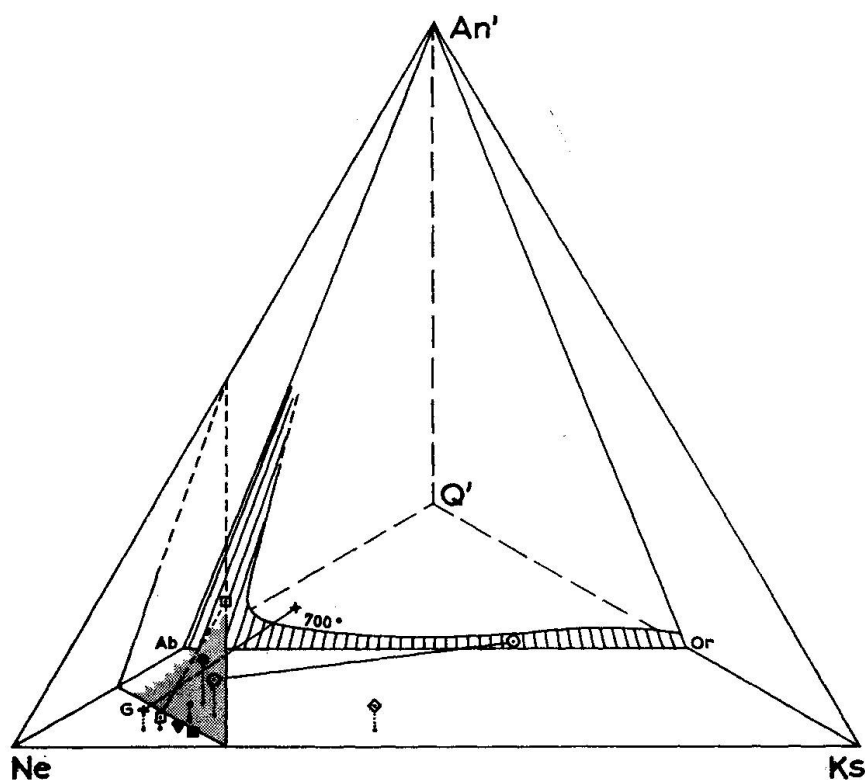Methuen, Ontario, TILLEY 1952.

Fig. 2. The quarternary diagram Ne-Ks-An'-Q'. The plane of the feldspar com-
position is indicated, Ab-Or-An'; the limits of solid solution in ternary feldspars
at 700° is indicated by hatching. The shaded plane is the plane of composition of
the low-temperature natural nephelines. Some nepheline and feldspar analyses are
shown. Coexisting pairs are joined by tie lines. Designation as in Fig. 1. (Note the
anomalous composition of G, which was also noticed by HAMILTON 1961 in an
other connection.)

composition of nephelines, that have crystallized from lavas. See point $\langle \cdot \rangle$ in Fig. 1.

Finally it is worth noting that a nepheline of essentially the composition of the pure sodium member, $NaAlSiO_4$, is not to be expected in nature. An inspection of Fig. 2 shows that the composition plane of natural nephelines separates this compound from quartz, all feldspars, leucite, and kalsilite, with which it, therefore, is incompatible.

Nepheline as a geologic thermometer

The previous discussion makes it clear that the composition of low-temperature nepheline is fixed by equations (1) and (2), and that in nature the composition is even more strictly limited because only small amounts of An and Q can go into solid solution (cp. the MOROZEWICS-HAHN-BUERGER formulas, and the compound $Na_3KAl_4Si_4O_{16}$ in the simple binary Ne-Ks system). It is also clear that, generally speaking, nephelines exhibiting higher contents of K_2O were formed at elevated temperature. But only in special cases is it possible to give the exact relations. In rocks containing both nepheline and kalsilite (or leucite), the K-content of nepheline is a reliable temperature indicator; the quantitative data can be taken directly from the phase diagram of TUTTLE and SMITH. Such rocks have been described by SAHAMA in various papers. But very few rocks contain kalsilite. In most rocks nepheline is associated with feldspar, and it is then impossible to use the K_2O content of nepheline as a practical geological thermometer.

For such rocks another geologic thermometer was first indicated by HAMILTON and MACKENZIE (1960) and subsequently given in details by HAMILTON (1961): the solubility of SiO_2 in Na-K mixed crystals of nepheline. Two of HAMILTON's curves are plotted into Fig. 1. The limitation of this thermometer lies in the difficulties of obtaining accurate chemical analyses. In order to calculate excess silica in nepheline, not only must SiO_2 itself be accurately determined, but also very accurate determination of Na_2O, K_2O, CaO and Al_2O_3 are necessary. Personally I do not know of any laboratory in which so reliable analyses may be performed.

Another possibility is to determine the solubility of calcium (as An = $CaAl_2Si_2O_8$) in the Na-K nephelines. BOWEN (1912) has shown that at high temperatures up to 35 weight per cent An may enter into the nepheline lattice. But no further measurements have been made,

and the petrological application might always be difficult. In order for
natural nepheline to take the maximum amount of Ca into solid solution
it should be in contact with pure calcium silicates, e. g. anorthite, and
this is very rarely the case. Rather the thermometer had to be based
on the partition of calcium between nepheline and feldspar. This is an
idea which BOWEN (1917) actually mentioned.

Table 5 shows this partition as found in various nepheline-feldspar
associations. It varies from zero to infinity, and can therefore not be
used. The reason is obviously that in nepheline-plagioclase associations,
most of the Ca goes into feldspar, while in nepheline-orthoclase associa-
tions it preferably enters into nepheline (see Fig. 2).

Table 5. *Calcium contents in associated pairs of nepheline and feldspars*

	1	2	3	4	A	B	C	D	E	F	G
An in nepheline, x_1	6.0	1.5	0	1.2	2.5	2.1	2.4	3.0	0.6	4	2.7
An in feldspar, x_2	0.6	7.7	4.9	4.0	0.6	2.3	2.5	1.7	0.0	1.6	6.0
x_1/x_2	1.0	0.2	0	0.3	4	0.9	1	2	∞	2.5	0.5
Or:Ab in feldspar	66:34	7:93	6:94	24:76	38:62	46:54	48:52	51:49	98:2	97:3	20:80

The best thermometer seems to me to be the tie lines in the system
Ab-Or-Ks-Ne (Fig. 1). They have also been used by TILLEY to distinguish
between various nepheline parageneses. HAMILTON and SMITH have
determined these tie lines at 700°. It is clear from the figure that at
this temperature the lines trend from the bottom obliquely upward to
the top of the diagram, while at lower temperatures the trend is more
straight upwards. Thus Fig. 1 shows that the nephelines of the lardalite
of southern Norway crystallized lower than 700°, but higher than those
from pegmatites of the Stjernö-Seiland area of northern Norway. De-
terminations of the trend of the lines at various temperatures would
probably be the best geological thermometer in nepheline-feldspar rocks.

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