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DIFFERENTIATION OF MELILITE NEPHELINITIC ROCKS  
IN LIGHT OF EXPERIMENTAL STUDY OF THE SYSTEM  
 $\text{Na}_2\text{O}-\text{CaO}-\text{MgO}-\text{Al}_2\text{O}_3-\text{TiO}_2-\text{SiO}_2$

by

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(with 1 table and 9 text-figures)

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*Abstract*

Differentiation of the melilite nephelinitic rock series is discussed in light of recent experimental data of the system  $\text{Na}_2\text{O}-\text{CaO}-\text{MgO}-\text{Al}_2\text{O}_3-\text{TiO}_2-\text{SiO}_2$ . The crystallization sequence in the join diopside-akermanite-nepheline- $\text{CaTiAl}_2\text{O}_6$  including principal constituent minerals in melilite nephelinite is particularly referred to construct an inferred differentiation trend. The trend is olivine nephelinite  $\rightarrow$  olivine-melilite nephelinite  $\rightarrow$  perovskite-bearing olivine-melilite nephelinite or melilite nephelinite  $\rightarrow$  perovskite-bearing melilite nephelinite  $\rightarrow$  corundum and perovskite-bearing melilite nephelinite. When this inferred trend is compared with the observation in natural melilite nephelinitic rocks, it is evident that this differentiation trend is now valid to elucidate the formation of the melilite nephelinitic rock series. The relationship between titaniferous pyroxene and perovskite is also discussed with the data of the join diopside- $\text{CaTiAl}_2\text{O}_6-\text{SiO}_2$ .

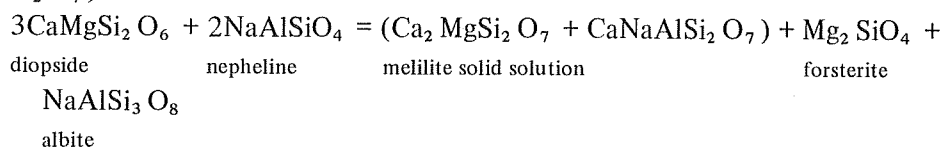
Introduction

The sodium-rich mafic and ultramafic volcanic rocks are characterized by higher  $\text{Na}_2\text{O}$  and  $\text{TiO}_2$  and lower  $\text{SiO}_2$  compared to rocks belonging to average basaltic rock series. The undersaturated character of these rocks are reflected by the presence of such minerals as nepheline, melilite, titanite and olivine. Some of the interesting rock types, belonging to this family include: nephelinite, melilite nephelinite, olivine-melilite nephelinite and olivine nephelinite, etc. The primary magma, responsible for the formation of these rocks, are believed by many petrologists to have been generated by partial melting of the upper mantle (Bultitude and Green, 1967, Green 1969, Kushiro, 1969, O'Hara, 1970, Bultitude and Green, 1971). However, the genesis and differentiation of these rock types under volcanic and sub-volcanic condition at shallow depths remain an interesting problem. Differentiation of these various rock type at 1 atm will be discussed in this paper in light of the experimental study of the system  $\text{Na}_2\text{O}-\text{CaO}-\text{MgO}-\text{Al}_2\text{O}_3-\text{TiO}_2-\text{SiO}_2$ .

The System  $\text{Na}_2\text{O}-\text{CaO}-\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2$ *Expanded basalt tetrahedron*

First experimental work on the melilite nephelinitic rocks has been carried out by Bowen (1922) who showed the preliminary phase equilibrium diagram of the system diopside-nepheline in which the principal constituent minerals of this rock type were encountered. Bowen also pointed out that it is of importance to study the system  $\text{CaO}-\text{MgO}-\text{SiO}_2$ -nepheline to elucidate the genesis and the differentiation of the melilite and nepheline-bearing rocks (Bowen, 1928, p. 164).

Later the system diopside-nepheline was studied in detail by Schairer et al. (1962) and olivine, clinopyroxene, melilite, and nepheline, all of which are main constituent minerals in the melilite nephelinitic rocks, were confirmed in the system. Olivine and melilite may be formed by the following reaction and melilite includes considerable amount of soda-melilite molecule ( $\text{CaNaAlSi}_2\text{O}_7$ ).



Schairer and Yoder (1964) studied the system nepheline-forsterite-larnite-silica including melilite and nepheline composition as a comprehensive model of basaltic rock. Since this is an expanded model of the basalt tetrahedra proposed by Yoder and Tilley (1962), it is called expanded basalt tetrahedron, which includes all the mineral assemblages observed in natural rocks.

The silica-undersaturated part of this system is related to the melilite nephelinitic rock series and the relationship among the mineral assemblages in the system and the rock terminology was given by Schairer and Yoder (1964) as follows: olivine nephelinite, olivine melilitite, olivine-melilite nephelinite, melilite nephelinite, wollastonite-melilite-nephelinite, wollastonite nephelinite, and wollastonite-nepheline tephrite. There are three invariant points in the silica-undersaturated part of the quaternary system; two are reaction points and one is a eutectic point. At one reaction point forsterite is consumed by the reaction with liquid, and at the other melilite is consumed. Such a phenomenon has not yet been described from natural rocks, but the reaction involving melilite is important for the incompatibility of melilite and plagioclase in natural rocks. Further petrographic data on these minerals are expected.

*The system diopside-akermanite-nepheline*

Bowen (1928) stressed the importance of the system  $\text{CaO}-\text{MgO}-\text{SiO}_2-$

nepheline. Onuma and Yagi (1967) studied the join diopside-akermanite-nepheline which is a part of this system. The liquidus diagram of the system diopside-akermanite-nepheline is shown in Fig. 1. The system has no invariant

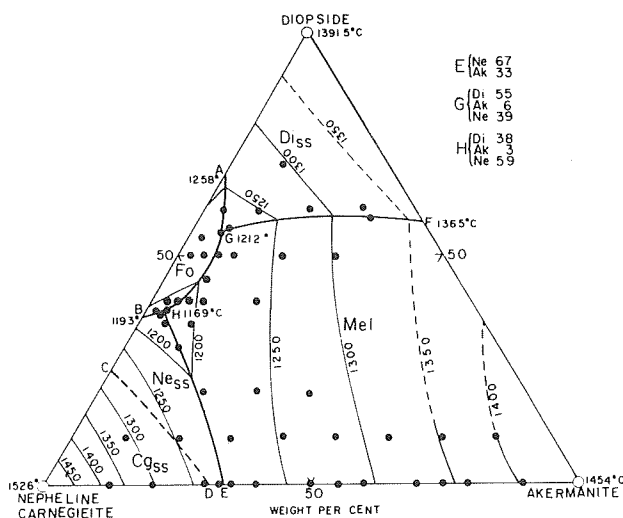


Fig. 1 Liquidus diagram for the system diopside-akermanite-nepheline (after Onuma and Yagi, 1967).  $Di_{SS}$  = diopside<sub>SS</sub>, Fo = forsterite, Mel = melilite,  $Ne_{SS}$  = nepheline<sub>SS</sub>,  $CG_{SS}$  = carnegieite<sub>SS</sub> (ss: solid solution).

point and contains two piercing points G and H. G lies on the univariant line separating the primary phase volumes of melilite, forsterite and diopside in the system CaO-MgO-SiO<sub>2</sub>-nepheline, and at this point forsterite, melilite, and diopside coexist with liquid. On the other hand H lies on the univariant line along which nepheline, forsterite, and melilite coexist with liquid. The position of these univariant lines in the system CaO-MgO-SiO<sub>2</sub>-nepheline is shown in Fig. 2. Both univariant lines proceed towards the plane of CaO-SiO<sub>2</sub>-nepheline with falling temperature. The fractional crystallization along these univariant lines produces a residual liquid rich in CaO and SiO<sub>2</sub>. The two univariant lines cross each other at an invariant point in the quaternary system and the liquid coexists with forsterite, diopside, melilite, and nepheline at this invariant point I as shown in Fig. 3. This is also confirmed by Schairer and Yoder (1964). After forsterite is consumed at this point, crystallization proceeds towards another invariant point with falling temperature. During this crystallization the residual liquid becomes enriched in CaO and SiO<sub>2</sub>, and finally wollastonite would crystallize out.

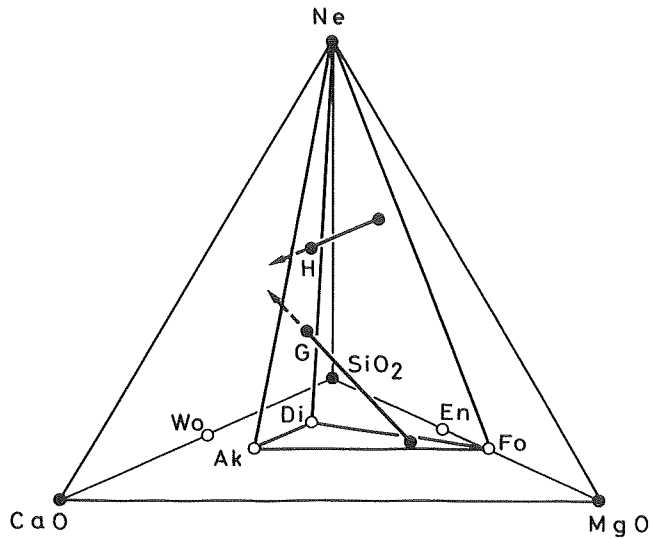


Fig. 2 Nepheline-CaO-MgO-SiO<sub>2</sub> tetrahedron showing two univariant lines and two piercing points in the join diopside-akermanite-nepheline.

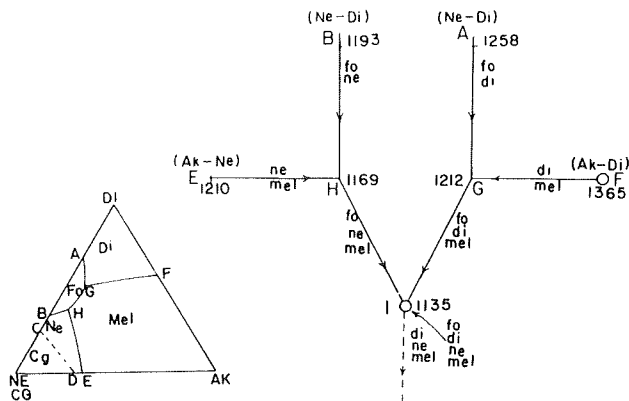
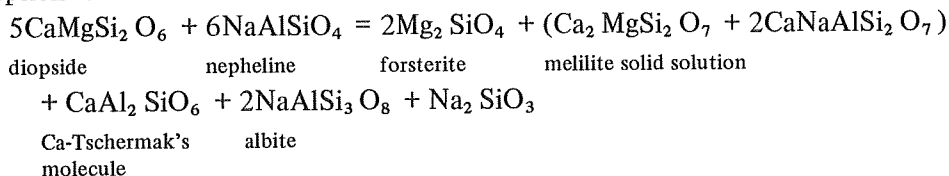


Fig. 3 Flow sheet of the system diopside-akermanite-nepheline showing an invariant point (reaction point) forsterite + diopside + nepheline + melilite + liquid (Onuma and Yagi, 1967).

In the system diopside-nepheline-akermanite the mineral assemblage at G corresponds to olivine melilitite and that at I to olivine-melilitite nephelinite. After forsterite disappears by reaction at I, the mineral assemblage represents that of melilite nephelinite. This system is a part of five component system but sometimes the crystallization ceases on a univariant line, resulting a three phase assemblage because of the complex solid solution of the phases encountered in

the system. For example, Onuma and Yagi (1967) have inferred that the diopside contains  $\text{CaAl}_2\text{SiO}_6$  (Ca-Tschermak's molecule) and melilite contains  $\text{CaNaAlSi}_2\text{O}_7$  (soda-melilite molecule) from the reaction between diopside and nepheline.



The melilites crystallized near G and H are nearly isotropic, showing that soda-melilite is incorporated. Yoder (1964) has shown that natural melilite contains 1/3 of soda melilite molecule.

#### *Possible differentiation course*

On the basis of the experimental results of Schairer and Yoder (1964) and Onuma and Yagi (1967), the differentiation course of the melilite nephelinitic rock series are inferred as shown in Fig. 4. The wollastonite in the last product

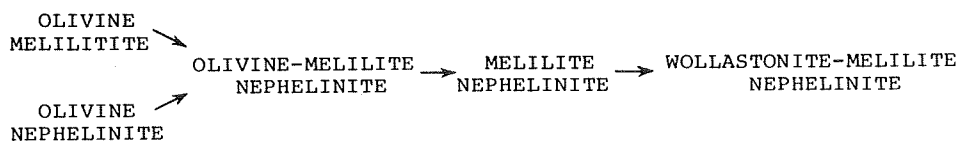


Fig. 4 Inferred differentiation course of the melilite nephelinitic rock series without taking  $\text{TiO}_2$  into account.

might be replaced by perovskite or sphene when the residual liquid is rich enough in  $\text{TiO}_2$  to produce one of these minerals. When the residual liquid is poor in  $\text{TiO}_2$ , however, it is incorporated in clinopyroxene, and wollastonite together with nepheline is consumed to form soda melilite, which in turn enters into the structure of akermanite to form solid solution (Schairer and Yoder, 1964). Thus neither perovskite nor sphene is crystallized.

As indicated in the above statements, the role of  $\text{TiO}_2$  in the melilite nephelinitic rock series is indeed very important in the differentiation of these rock series which is richer in  $\text{TiO}_2$  than tholeiite rock series and ordinary alkaline rock series.

The System  $\text{Na}_2\text{O-CaO-MgO-Al}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2$  and the Differentiation of

## Melilite Nephelinitic Rock Series

Yagi and Onuma (1969) expanded their work of 1967 and studied the system diopside-akermanite-nepheline- $\text{CaTiAl}_2\text{O}_6$  to elucidate the role of titanium in alkalic basalt, with special reference to the melilite nephelinitic rocks. The  $\text{CaTiAl}_2\text{O}_6$  molecule is significant as a constituent in clinopyroxenes in alkaline rocks (Kushiro, 1960, Yagi and Onuma, 1967). In the system diopside- $\text{CaTiAl}_2\text{O}_6$  the clinopyroxene incorporates  $\text{CaTiAl}_2\text{O}_6$  molecule as much as 11 wt. % and proskite coexists with titaniferous clinopyroxene, melilite, and spinel at subsolidus temperatures in the  $\text{CaTiAl}_2\text{O}_6$ -rich portion.

Perovskite has been reported from natural olivine-melilite nephelinite (Winchell, 1947, and Spenser, 1969) and the clinopyroxenes in these rocks are rich in  $\text{CaTiAl}_2\text{O}_6$ . The system diopside-akermanite-nepheline- $\text{CaTiAl}_2\text{O}_6$  is thus an important model for the genesis of the melilite nephelinitic rocks. This system is a part of the six-component system  $\text{Na}_2\text{O}-\text{CaO}-\text{MgO}-\text{Al}_2\text{O}_3-\text{TiO}_2-\text{SiO}_2$ . However, the number of the phases never exceeds 6 and their melting interval is rather small (less than  $35^\circ\text{C}$ ) and consequently the system behaves like a five-component system. In this case a five-phase assemblage can be treated as a univariant line and six-phase assemblage as an invariant point.

The tetrahedron of the system diopside-akermanite-nepheline- $\text{CaTiAl}_2\text{O}_6$  is shown in Fig. 5. Eight five-phase assemblages and four six-phase assemblages were confirmed in the system. Fig. 6 show the flow sheet constructed from

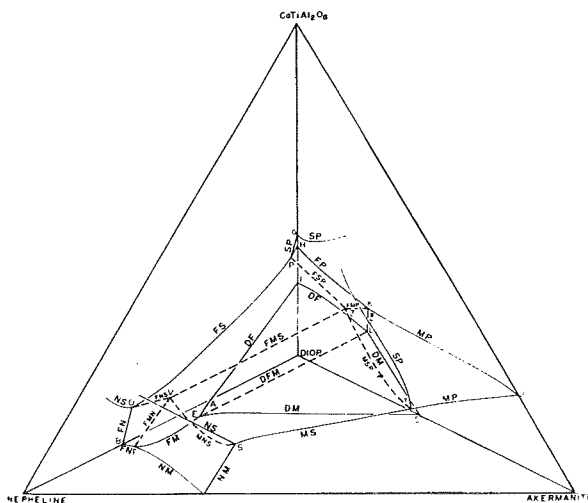


Fig. 5 Tetrahedron representing liquidus phase volumes in the system diopside-akermanite-nepheline- $\text{CaTiAl}_2\text{O}_6$  (after Yagi and Onuma, 1969). D = diopside, F = forsterite, M = melilite, N = nepheline, P = perovskite, S = spinel.

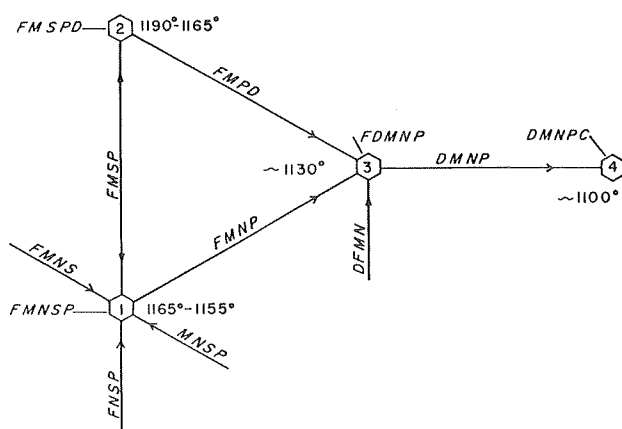


Fig. 6 Mutual relations of five-phase and six-phase assemblages in the system diopside-akermanite-nepheline- $\text{CaTiAl}_2\text{O}_6$  (after Yagi and Onuma, 1969). C = corundum and remainder abbreviations are same as in Fig. 5.

these experimental results. At V and Z in Fig. 5 both showing five phase assemblages liquids coexist with forsterite, spinel, melilite, and nepheline and with forsterite, spinel, melilite, and perovskite, respectively.

Natural rocks, corresponding to mineral assemblages 1, and 2 of Fig. 6 have not been found. The phase assemblage of DFMN (diopside + forsterite + melilite + nepheline + liquid) corresponds to olivine-melilite nephelinite and the assemblage at 3 corresponds to perovskite-bearing olivine-melilite nephelinite. At 3 forsterite is consumed and the assemblage between 3 and 4 indicates perovskite-bearing melilite nephelinite. The temperature falls towards 4 where corundum crystallizes out.

As mentioned above, the crystallization course which proceeds along DFMN through point 3 and along DMNP ultimately to 4 has a special petrologic significance. It is therefore interesting to analyze relationship between DFMN and the four-phase assemblage which is generated from the five-phase assemblage along DFMN. These five-phase assemblages were also confirmed as invariant points in the system diopside-akermanite-nepheline. In Fig. 7 a schematic flow diagram is given. The temperature at which a four-phase assemblage (three crystalline phases + liquid) transfers to a five-phase assemblage is not fixed and consequently DFMN is a reaction line along which forsterite reacts with liquid with falling temperature. In Table 1 crystallization sequences of selected mixtures in the system diopside-akermanite-nepheline- $\text{CaTiAl}_2\text{O}_6$  are given with the bulk composition of starting material. The data show that the crystallization sometimes ceases on the univariant line or divariant plane before the eutectic point is reached, because of the complex solid solution of



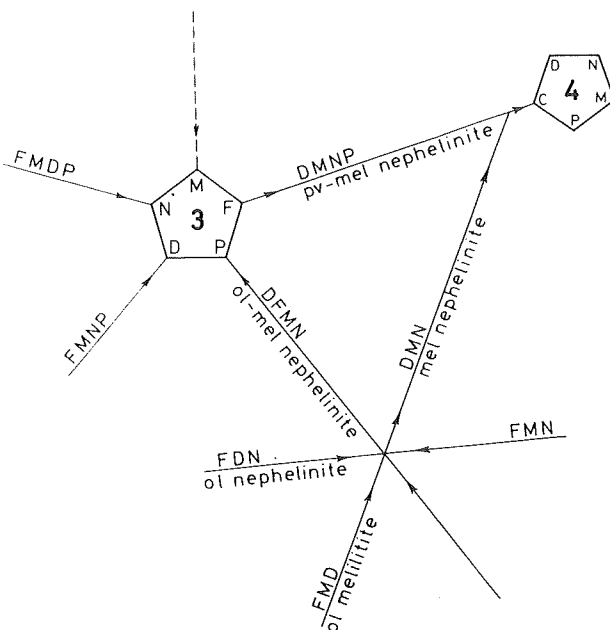


Fig. 7 Mutual relation of invariant, univariant, and divariant assemblages of a part of the system  $\text{Na}_2\text{O}-\text{CaO}-\text{MgO}-\text{Al}_2\text{O}_3-\text{TiO}_2-\text{SiO}_2$  with corresponding rock names. Abbreviations are same as in Figs. 5 and 6.

the crystalline phases. On the basis of these data it is evident that if the liquid is rich in  $\text{TiO}_2$  and poor in  $\text{SiO}_2$  the crystallization proceeds along the DFMN until point 3 is reached, where perovskite joins the assemblage, while if the liquid is poor in  $\text{TiO}_2$  and rich in  $\text{SiO}_2$ , forsterite is consumed by the reaction before point 3 is reached, and the crystallization course leaves the DFMN and proceeds along the DMN (actually divariant plane), separating melilite, nepheline, and diopside (melilite nephelinite).

O'Hara and Biggar (1969) suggested that the melilite nephelinite is formed by the low-pressure extreme fractional crystallization of alkali olivine basalt magma. Recently Onuma and Yamamoto (1976) also indicated that the crystallization at depth does not lead to the melilite nephelinite but produce the olivine nephelinite and that the olivine melilitite and the melilite nephelinite are formed by the low pressure fractional crystallization of the magma. Considering these two discussions, it seems that the crystallization sequence obtained by the experimental data at atmospheric pressure is valid for the interpretation in the differentiation of the natural melilite nephelinitic rock series.

From the above discussions an inferred crystallization differentiation of the

**Table 1** Crystallization sequence of the selected compositions in the system diopside-akermanite-nepheline-CaTiAl<sub>2</sub>O<sub>6</sub>.  
(data are quoted from Yagi and Onuma, 1969)

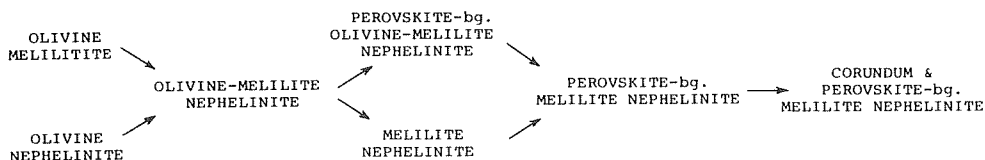
|     | Di | Ak | Ne | Tp | Crystallization Sequence  |
|-----|----|----|----|----|---|
| (1) | 40 | 10 | 40 | 10 | L → Fo+L → Fo+Mel+L → Fo+Mel+Ne+L → Mel+Ne+Di+Pv+L → Mel+Ne+Di+Pv+Cor             |
| (2) | 75 | 5  | 5  | 15 | L → Fo+L → Fo+Di+L → Fo+Di+Mel+L → Fo+Di+Mel+Pv+L → Di+Mel+Pv+Ne+L → Di+Mel+Pv+Ne |
| (3) | 70 | 10 | 10 | 10 | L → Fo+L → Fo+Di+L → Fo+Di+Mel+L → Fo+Di+Mel+Ne+L → Di+Mel+Ne+Pv+L → Di+Mel+Ne+Pv |
| (4) | 70 | 10 | 15 | 5  | L → Fo+L → Fo+Mel+L → Fo+Mel+Di+L → Mel+Di+Ne+L → Mel+Di+Ne                       |

|                                | (1)   | (2)   | (3)   | (4)   |
|--------------------------------|-------|-------|-------|-------|
| SiO <sub>2</sub>               | 43.73 | 45.96 | 47.54 | 49.61 |
| TiO <sub>2</sub>               | 3.36  | 5.04  | 3.36  | 1.68  |
| Al <sub>2</sub> O <sub>3</sub> | 18.65 | 8.24  | 7.88  | 7.54  |
| MgO                            | 8.92  | 14.69 | 14.50 | 14.50 |
| CaO                            | 16.82 | 25.02 | 24.59 | 23.42 |
| Na <sub>2</sub> O              | 8.72  | 1.09  | 2.18  | 3.27  |

Di = diopside (CaMgSi<sub>2</sub>O<sub>6</sub>), Ak = akermanite (Ca<sub>2</sub>MgSi<sub>2</sub>O<sub>7</sub>), Ne = nepheline (NaAlSi<sub>3</sub>O<sub>8</sub>), Tp = CaTiAl<sub>2</sub>O<sub>6</sub>, Fo = forsterite, Pv = perovskite, Mel = melilite, Cor = corundum, L = liquid.

melilite nephelinitic rock series is as shown in Fig. 8. Now the wollastonite in the wollastonite-melilite nephelinite is replaced by perovskite, supporting a suggestion by Schairer and Yoder (1964). Perovskite was reported from the natural olivine-melilite nephelinite and melilite nephelinite by Winchell (1947) and Spencer (1969). Spencer also described that perovskite is observed in the olivine-melilite nephelinite but not in the olivine nephelinite. This would support the inferred differentiation course of the melilite nephelinite rock series.

Although corundum has not been reported as a constituent of melilite



**Fig. 8** Inferred differentiation course of the melilite nephelinitic rock series with taking TiO<sub>2</sub> into account.

nephelinite, occurrence of corundum megacrysts in nepheline basanite, hawaiite or related alkalic rocks from several localities in northern Queensland, Australia is worthy of notice (Stephenson, 1976). At Cheviot Hills, for example, rounded corundum crystals, sometimes up to 10 mm in diameter, are found associated with abundant, corroded megacrysts of anorthoclase. Genetic relation between the corundum and the host rocks has not yet been clarified, and the origin from the deep buried metamorphic rocks cannot be denied. It is, however, also possible to consider that corundum might have grown directly from the nepheline normative basaltic melt in the later stage under some special conditions, as supported by the results of the recent experiments.

Recently we had a chance to observe the olivine-melilite nephelinites from Eifel, West Germany. The rocks contain phenocrysts of olivine and titaniferous clinopyroxene and the groundmass always carries clinopyroxene, nepheline, melilite and perovskite (Detailed study on this rock will be published elsewhere by Yagi et al.). It is noticed that the phenocrystic olivine has corroded anhedral form and that olivine is absent and perovskite is always present in the groundmass. This observation leads to the following process during the formation of olivine-melilite nephelinite. The magma crystallizing olivine and clinopyroxene both of which are observed as phenocrysts (A-G in Fig. 3) was erupted. Then the crystallization course followed FMD and DFMN (Fig. 7), precipitating clinopyroxene, melilite, and nepheline, and finally reached the point 3, where perovskite joined in the groundmass. During this process olivine did not precipitate but decreased in amount by a reaction with liquid as experimental data showed. In nature, generally crystallization ceased at this point before the olivine is completely consumed, resulting in the formation of melilite nephelinitic rock always carrying olivine. Probably, the magma was cooled too quickly to follow further crystallization, because the most part of crystallization took place after the magma was erupted.

When the liquid is rich in  $\text{SiO}_2$  sphene crystallizes instead of perovskite and the mineral assemblages would be different. The relationship between sphene and perovskite is controlled by activity of silica as will be stated in the following section.

#### Relation Between Perovskite and Sphene and the System $\text{CaO-MgO-Al}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2$

Gupta et al. (1973) studied the system diopside- $\text{CaTiAl}_2\text{O}_6$ -silica. In this system there is an interesting relationship between pyroxene and associated titanium-bearing minerals, sphene and perovskite. The flow sheet given in Fig. 9 indicates that silica content increases from the upper part to the low part. Two

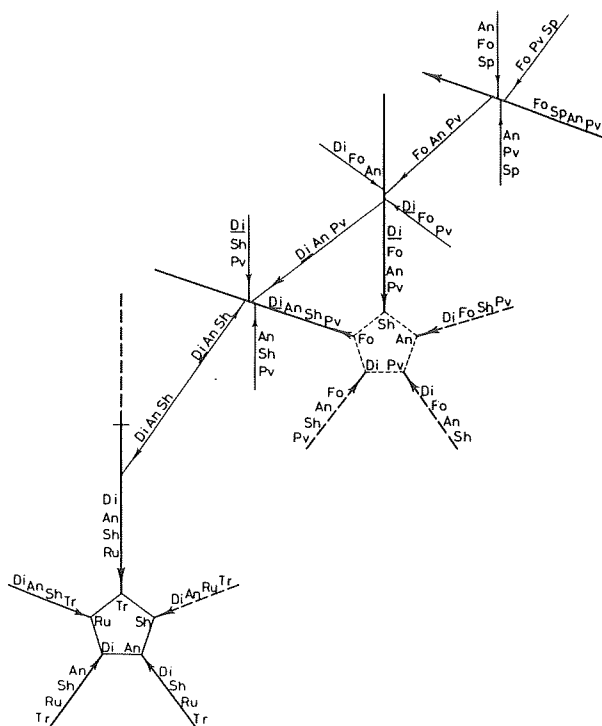
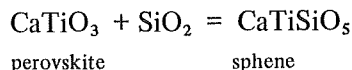


Fig. 9 Flow sheet of the system diopside- $\text{CaTiAl}_2\text{O}_6$ -silica (after Gupta et al.).

kinds of clinopyroxene were obtained in the system. One is almost pure diopside and the other titanium-bearing clinopyroxene solid solution in the silica-poor portion, while sphene with pure diopside in the silica-rich portion. When perovskite and sphene coexist the associated clinopyroxene is pure diopside. Further, sphene does not coexist with forsterite. Smith (1970) pointed out that forsterite coexists with perovskite but not with sphene, in the rocks rich in titanium and undersaturated with silica. This observation is consistent with the experimental results.

From the fact that perovskite is rarely found with feldspar, Carmichael et al. (1970) indicated that the following reaction sets a lower limit of silica activity in feldspar-bearing rocks:



However, in the system diopside- $\text{CaTiAl}_2\text{O}_6$ -silica perovskite does coexist with anorthite. This fact is inconsistent with the observation in natural rocks, and may probably be ascribed to chemical composition of the starting liquid. In the system  $\text{CaO-MgO-Al}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2$  some starting liquids in the silica-rich portion produce the phase-assemblage including perovskite and anorthite, while

anorthite may be replaced by melilite in the silica-poor portion, and thus the assemblage may correspond to the natural assemblage (cf. olivine melilitite).

In the system diopside-akermanite- $\text{CaTiAl}_2\text{O}_6$  representing the silica-poor portion of the system  $\text{CaO-MgO-Al}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2$  Onuma and Yagi (1971) found the phase assemblage of diopside + forsterite + akermanite + perovskite + liquid (olivine melilitite) instead of diopside + forsterite + anorthite + perovskite + liquid which is encountered in the system diopside- $\text{CaTiAl}_2\text{O}_6$ -silica. These two joins may be connected by the divariant assemblage of forsterite + diopside + perovskite + liquid.

When sodium is added to the system, nepheline instead of pure anorthite, would appear in the silica-undersaturated portion, whereas albite-rich plagioclase would appear in the silica-rich portion of the system  $\text{Na}_2\text{O-CaO-MgO-Al}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2$ . O'Hara and Biggar (1969) and Schairer and Yoder (1964) reported that in the system  $\text{CaO-MgO-Al}_2\text{O}_3\text{-SiO}_2$  anorthite and melilitite coexist, showing inconsistency with the observation in natural rocks. However, the addition of  $\text{Na}_2\text{O}$  to the system changes the phase-assemblage and melilitite never coexist with plagioclase in the system  $\text{Na}_2\text{O-CaO-Al}_2\text{O}_3\text{-SiO}_2$ , indicating that a increase of component makes the system closer to the natural rocks.

#### *Acknowledgement*

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