

The GEOCHEMISTRY of the OLIVINE MELILITES and RELATED
ROCKS of NAMAQUALAND-BUSHMANLAND, SOUTH AFRICA

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"And some rin up hill and down dale
knapping the chunky stanes to pieces
wi' hammers, like sae many road makers
run daft. They say it is to see how
the world was made."

SIR WALTER SCOTT



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ABSTRACT

Early Tertiary olivine melilitites and associated non-diamondiferous kimberlites occur in two pipe clusters in the Namaqualand and Bushmanland regions of South Africa. The more northerly cluster is centred about the town of Gamoep and the second is located south of Garies.

The olivine melilitites are characterized by olivine phenocrysts which exhibit a broad range in composition on the scale of individual hand specimens. It is possible to distinguish four petrographically and chemically distinct olivine populations in both the northern and southern pipes studied:

- (a) Rare anhedral or subhedral olivines that display marked disequilibrium features with the surrounding matrix and which are characterized by having high iron and extremely low nickel contents (referred to as HILN olivines) relative to the remaining olivines in the same rock;
- (b) euhedral and often skeletal (hopper) olivines that are richer in Mg and Ni than HILN olivines in the same rock. Some of the euhedral olivines show abnormal zonation patterns ("unusual" hoppers). Hopper and HILN olivines contain fluid and carbonate inclusions which apparently record the separation of a vapour phase and an immiscible carbonate liquid during magma ascent;
- (c) large rounded olivines (megacrysts) up to 40 mm across. Individuals are chemically homogeneous, but megacrysts from the same pipe collectively define a trend of decreasing Mg with Ni (Fo_{92} , 0.36% Ni to Fo_{75} , 0.17% Ni). The most fayalitic megacrysts are depleted in Mg and Ni relative to the hopper olivines in the same rock.
- (d) In addition to these main populations, there also occur rare magnesium-rich (Fo_{91}) anhedral olivines which show strained extinction and are believed to be xenocrysts.

It is suggested that the HILN olivines crystallized from primitive carbonate-rich magmas under conditions of low oxygen fugacity, intermediate between the Ni-NiO and Fe-FeO buffers. Mineral-melt partition coefficients for the transition elements determined in basaltic systems are considered to be inappropriate to such carbonate-rich melts. Separation of volatiles and an immiscible carbonate liquid during magma ascent resulted in an increase in oxygen activity, a decrease in the Fe-Mg distribution coefficient (K_D) for olivine and residual silicate melt, and an increase in liquidus temperatures. These effects led to the rapid crystallization of Mg- and Ni-enriched skeletal hopper olivines. The "unusual" hopper olivines crystallized later than the HILN olivines but prior to the "normal" hoppers under conditions characterized by rapid and independent changes in oxygen activity and partition coefficients associated with the loss of volatiles and an immiscible carbonate liquid. The range in chemistry which characterizes the megacryst olivine suite is believed to record physico-chemical changes in the magmas subsequent to separation from the mantle source area, but prior to crystallization of the HILN olivines. Most important of these changes was an increasing degree of polymerization of the liquid structure and a progressive decrease in oxygen activity as the molar ratio ($X_{CO_3''}/(X_{CO_3''} + X_{CO_2})$) in the magma increased with decreasing confining pressure. Increasing polymerization of the liquid resulted in an increase in olivine-liquid partition coefficients for transition elements.

Olivine melilitite bulk rock compositions are characterized by a wide range in chemistry, and by a marked decoupling of the geochemical behaviour of the "compatible" and "incompatible" elements. The northern pipes are enriched in Mg, Si, Ni, Co and Cr but depleted in Fe, Ca, Al, P and Y and markedly depleted in Sr, Nb and Zr relative to the pipes in the south. K, Rb and Ba concentrations show no systematic variation across this compositional spectrum.

Bulk rock chemical variation in the Gamoep pipes was dominated by addition and subtraction of high pressure mineral phases (olivine, clinopyroxene and ilmenite), while low pressure fractionation involving olivine, melilitite, titanomagnetite and perovskite appears to have determined chemical variation in the Garies pipes. Source area inhomogeneity, and loss of a vapour phase and immiscible carbonate liquid during magma ascent played a secondary role in modifying bulk rock chemistry within each pipe cluster.

Major element variation between the two pipe clusters can be explained in terms of high pressure mineral fractionation involving olivine, clinopyroxene and ilmenite. However variations in incompatible trace elements are inconsistent with a simple fractionation model, and may in part be related to source area inhomogeneity and differences in the degree of partial melting.

Mineral chemistry data indicate that the parent magmas to kimberlites followed complex paths of physico-chemical evolution closely analogous to those inferred for the olivine melilitites, suggesting a close genetic link between the two rock types. Kimberlites are in general enriched in Mg and depleted in several incompatible elements relative to the olivine melilitites and are interpreted as being relatively less evolved rocks than the latter. Mineralogical differences between the two rock types are ascribed to the efficient separation of CO_2 and an immiscible carbonate liquid from the olivine melilitites, and inefficient separation of these components from kimberlites.

There is a close temporal relationship between episodes of alkaline volcanism and epeirogenic upwarping in southern Africa. Volcanic activity was triggered by stresses associated with such upwarping.

1. INTRODUCTION

1.1 GENERAL

Rocks which are loosely termed basalts display a wide spectrum of chemical variation, and occur in a wide variety of geological environments. They would appear to have in common a mantle origin, and an understanding of the genetic interrelationships of members of the basalt clan is clearly fundamental to the understanding of mantle processes, and the evolution of basic igneous rocks.

Recently, igneous petrologists have devoted much energy to the study of kimberlites, which may perhaps be regarded as constituting an alkaline ultra-basic extension of the basalt chemical spectrum. The presence of diamonds in many kimberlites indicates that these magmas are of relatively deep mantle origin and they possibly represent the deepest derived of all magma compositions. The kimberlite xenolith suite has received particular attention, for many of the accidental inclusions are interpreted as being samples of mantle material. However, the kimberlite host has been comparatively neglected, being inherently difficult to study as primary minerals are invariably affected by secondary alteration, while the almost ubiquitous xenoliths make contamination of these magmas a serious problem. Several authors (e.g. Taljaard, 1936; Holmes, 1936 and Yoder, 1975) have suggested that there is a close genetic relationship between kimberlites and olivine melilitites, indicating that a study of the latter may have bearing on deep-seated mantle processes in general and on the

origin of kimberlites in particular. These suggestions are supported by a close spatial association of kimberlites and olivine melilitites in Siberia (Ukhanov, 1963). Such an association is also found in the Namaqualand-Bushmanland region of southern Africa (see Fig. 1.1 for locality), and this provided the incentive for the present geochemical study. The project has been aimed primarily at rationalizing the origin of the olivine melilitite suite, but it was hoped that an understanding of the geochemistry of these volcanics would throw some light on the possible genetic relationship to the associated kimberlites, and, more ambitiously, other alkali-ne volcanics elsewhere in southern Africa.

1.2 LOCALITY

The Namaqualand-Bushmanland volcanics occur as small eroded pipes (described in Section 2) that define two distinct clusters. The larger of these is centred about the village of Gamoep and extends NNE to Aggeneys, while the second is located just to the south of the village of Garies (Fig. 1.1). Localities of individual pipes are given in Table 7.

Gamoep is situated close to the western edge of the Bushmanland plateau, which slopes gently eastwards, and is interpreted by King (1963) as representing a Miocene erosion surface. The Namiesberg to the north rises some 150 to 200 metres above this plateau, and their flat tops are considered to represent the remnants of an ancient pediplain, described as the Namaqualand Highland surface (Mabbutt, 1955). Fossil dinosaur bones in the Henkries area indicate a Cretaceous age for this

earlier surface.

West of the escarpment, and stretching to the Atlantic Ocean is a younger erosion surface of probable Pliocene age (King, 1972) that forms the region known as Namaqualand. This younger surface includes the village of Garies and the pipe cluster to the south of that village.

The Bushmanland-Namaqualand region is semi-arid, and the population consequently sparse and mainly occupied by karakul and goat farming, although mining operations are assuming increasing importance in the north of the region. Copper has long been recovered from enigmatic "noritoid" bodies in the Springbok area, while more recently, large stratiform copper-lead-zinc deposits have been discovered near Aggeneys and the Gamsberg. Alluvial diamonds are recovered from coastal gravels on either side of the Orange River mouth, and also from many of the river courses to the north as far as Benguela in Angola, and from those to the south as far as the Olifants River. The primary source of these diamonds remains unknown, but would appear to be unrelated to the Namaqualand-Bushmanland kimberlites, for these pipes have been extensively prospected and are apparently undiamondiferous (H Jenner-Clarke, oral communication). Nevertheless, active diamond prospecting in the area is still in progress and Cornelissen and Verwoerd (1975) suggest that this may yet prove to be rewarding. High quality marble is quarried from Nama sediments to the south of Van Rhynsdorp. Monazite has been recovered from the Steenkampskraal mine, though this is no longer operation-

al. Further mining activity in the region is restricted to sporadic small-scale exploitation of pegmatite bodies that are genetically related to the 1 000 m.y. Namaqualand gneisses.

1.3 PREVIOUS WORK

Rogers (1911) provides the earliest field and petrographic descriptions of the olivine melilitites of the Bushmanland plateau, and credits a Mr J G W Leipoldt - a surveyor in the area - as being the first person to recognize their volcanic character. Rogers (op. cit.) drew attention to the similarity between these pipes and those in the Sutherland district which had previously been described by Rogers and Du Toit (1903, 1904). The latter authors recognized that the olivine melilitites "*are all of a peculiar type and are more closely allied to the Kimberley pipes than to the more usual volcanic vents that are represented... by the Stormberg necks*".

Reuning (1931) first reported the presence of sediment-filled pipes on the Bushmanland plateau to the south of Gamoep, and concluded that these were kimberlites. Descriptions of the sedimentary infill and associated fossils in these pipes are provided by Haughton (1931), Rennie (1931) and Reuning (1934). Haughton (op. cit.) tentatively suggested that the pipes might be of late Cretaceous to early Tertiary age on the basis of circumstantial and rather equivocal fossil evidence.

The first comprehensive study of all known South African olivine melilitites was undertaken by Taljaard (1936). Apart from the meticulous petrographic descriptions, this study is most

noteworthy for the suggestion that kimberlites are closely allied to olivine melilitites, and might in fact be simply hydrothermally altered equivalents of the latter. On the basis of field relationships, Taljaard suggested that the olivine melilitites of the Sutherland commonage had been emplaced as three sills. This author further proposed that the Bushmanland olivine melilitites might be remnants of a previously extensive sill that spread out along the contact between the Namaqualand gneiss and the inferred overlying Dwyka sediments. Gerrard (1958) has partially supported Taljaard's interpretation of field relationships on the Sutherland commonage, but there has been no support for the interpretation of the Bushmanland olivine melilitites as remnants of an eroded sill.

Holmes (1936) in a review of "kimberlites and its associates" supported arguments for a strong genetic relationship between kimberlites and olivine melilitites. Although cautious not to make too explicit suggestions regarding the details of such a link, he suggested that kimberlites might be regarded as being composite mixtures of a magmatic component, similar in chemistry to olivine melilitites, volatiles such as H_2O and CO_2 , and xenolithic peridotites.

Subsequent to these early studies, South African olivine melilitites were comparatively neglected until Gerrard (1958) made a comparative mineralogical study of the Sutherland, Spiegel River and Klaasvoogds pipes.

Jansen (1960) reported the presence of olivine melilitite plugs

at Biesiesfontein to the south of Garies.

Moore (1973) presented a preliminary interpretation of bulk rock chemical data for the Namaqualand olivine melilitites.

Cornelissen and Verwoerd (1973, 1975) provided field descriptions of a number of Bushmanland diatremes and drew attention to local structural controls on their distribution. These authors summarized unpublished chemical analyses for the Bushmanland volcanics, and concluded that a number of pipes were true kimberlites on the basis of the associated heavy mineral suite.

Moore (1976) attempted to show that post-Gondwanaland alkaline volcanism in southern Africa has been largely tectonically controlled. Emplacement of the Namaqualand-Bushmanland pipes was ascribed to epeirogenic uplift of the continent.

Moore and Erlank (1977) drew attention to complex olivine zonation patterns in the Namaqualand-Bushmanland olivine melilitites. Such zonation was in part ascribed to large variations in oxygen activity during magma evolution.

In addition to the above studies, Mr H Jenner-Clarke of Assam Minerals, Springbok has for several years been carrying out an extensive prospecting programme in the Namaqualand-Bushmanland region, and has provided the author with valuable information on the olivine melilitites and associated diatremes in the area.

2. FIELD RELATIONSHIPS

On the basis of superficial field similarities, it is possible to distinguish several different varieties of volcanic pipes in the Gamoep and Garies areas. These will be referred to as:

- (1) Olivine melilitite and olivine nephelinite pipes
- (2) "Pseudo-Kimberlites"
- (3) Sediment-filled (Kimberlite) pipes and breccia necks.

2.1 OLIVINE MELILITITE AND OLIVINE NEPHELINITE PIPES

2.1.1 GENERAL

The olivine melilitites and olivine nephelinites occur as small eroded plugs that range in size from a maximum of roughly 400 m to less than a few tens of metres in greatest diameter (Plates 1 and 2). Some of the pipes show a poorly developed elongation parallel to the strike of the surrounding country rock, which in all cases is the 1 000 m.y. Namaqualand gneiss. Exposures are generally poor, and frequently restricted to a collection of boulders scattered about the surface of poorly defined depressions or pans or as rubble-strewn mounds or low hills (koppies). Occasionally, poorly developed columnar jointing, dipping outwards at low angles, is preserved at the pipe margins. This presumably results from rapid chilling of the magma against the country rocks. Isolated pegmatitic boulders (described in Section 3.3) are found in many of the pipes. Actual outcrops of such rocks have however only been found on the Zwartheuwel and Hoedkop pipes, where they occur as small irregular segregations a few metres across. It is estimated that these pegmati-

tes constitute appreciably less than 1% of the volume of the pipes in all cases.

2.1.2 HOEDKOP PIPE

Field relationships are best preserved in the Hoedkop pipe, which is a relatively large, slightly elongate plug some 200 m in greatest diameter. This pipe is therefore described in more detail. The contact with the country rock gneiss is somewhat irregular (Plate 1) but judging from the limited exposure, appears to be sharp. There is no evidence of fenitization of the country rocks, although in one restricted zone a few metres across and close to the margin, biotite in the gneiss has been altered to chlorite, feldspars are kaolinized and secondary chalcedony is developed.

At the eastern margin of the Hoedkop pipe, there is a coarse, roughly vertical banding parallel to the contact with the country rocks (Plate 3). This banding reflects the presence of irregular, phlogopite-rich schlieren that are somewhat less resistant to alteration, and have been selectively etched out by weathering. The banding is accompanied by small scale flow structures (Plate 4) and is therefore interpreted as a larger scale flow feature. Shale fragments with bleached margins are concentrated about the edge of the pipe. These must presumably have been carried downwards from a stratigraphic level above the country rock gneiss. Some of the higher hills in the vicinity of the Hoedkop pipe have a capping of Dwyka (lower Karroo) shales and such sediments would appear to be a likely source

for the shale inclusions. This evidence, together with the flow structures, indicates a marginal downflow of magma at the time of emplacement, analogous to that invoked by Philippotts (1968) to account for vertical igneous layering in the Mount Johnson pipe. Boulders showing flow structures have only been recovered from one other pipe (Bruinkop A), and in view of the poor field exposures, it is uncertain whether or not all were characterized by a marginal downflow of magma during emplacement.

The Hoedkop pipe is unusual in having a large number of different types of inclusions. These are found throughout the pipe, though appear to be more concentrated in restricted areas close to the margins. Apart from the shale fragments described above, the most important varieties recognized are:

- (a) Fragments of gneiss and granulite that were presumably derived from the surrounding country rocks or from lower crustal levels.
- (b) Rounded ultramafic inclusions, many of which are presumed to be of upper mantle derivation. Most common of these are spinel harzburgites, and olivine megacrysts, though very rare garnet lherzolites are also present. A spinel-garnet lherzolite with very pale mauve garnets was also found, though this unfortunately disintegrated during attempts to recover it from the extremely hard enclosing matrix. Spinel harzburgites and olivine megacrysts similar to those found at Hoedkop are also fairly abundant in the Zwartheuwel pipe, though are rare in other

Gamoep pipes and have not been found in any of the Garies pipes. Garnet lherzolites appear to be restricted to the Hoedkop pipe. Petrographic descriptions of the ultramafic nodules are given in Section 3.4. Analyses of the phases present in a garnet lherzolite (Ho-x) recovered from the Hoedkop pipe are presented in Table 2(o). These indicate equilibration at pressures of 25 Kb and temperatures of 937°C using the geothermometer and geobarometer developed by Fraser and Lawless (1978). As garnet lherzolite xenoliths are normally only reported from kimberlites, the recovery of such nodules from the Hoedkop pipe assumes special interest. Dr E M J Skinner of De Beers Consolidated Mines (oral communication, 1978) suggested on mineralogical grounds that this pipe might be a kimberlite, although in terms of bulk rock chemistry, the Hoedkop rocks that have been analysed are clearly related to the Namaqualand olivine melilitites (Section 4).

2.1.3 ZANDKOPSDRIEF PIPE

The Zandkopsdrift pipe, to the south of Garies, also displays a number of features that have bearing on the genesis of the olivine melilitites, and is therefore also described in some detail.

This pipe forms a low calcrete covered hill, several hundred metres across. Exposures are poor and the pipe margin ill defined, but a 10 m deep prospecting pit at the crest of the hill has produced a variety of different rock types. These include

one fresh olivine melilitite boulder, numerous fragments of altered olivine melilitite, often cemented together by carbonates, biotite-rich (glimmerite) nodules a few centimetres in diameter, and boulders with abundant phlogopite and ilmenite set in an earthy matrix. The latter superficially resemble some kimberlites, though apparently lack garnet and clinopyroxene xenocrysts. Close to the prospecting pit are two narrow (0.5 m) dykes that crop out continuously for a short distance downslope before being lost in the calcrete rubble. One is composed predominantly of carbonates, although a semi-quantitative analysis indicates a Ba content of up to 3.5%. This suggests a genetic relationship with the Salpetrekop carbonatite near Sutherland, which is characterized by having appreciable amounts of barite, and is spatially associated with a group of olivine melilitites (Rogers and Du Toit, 1903, 1904; Gerrard, 1958). The second dyke has an unusual banded appearance, due to the presence of zones of oolitic chalcedony alternating with zones rich in iron-manganese oxides. A specimen from this dyke showed a slight reaction with dilute acid, indicating the presence of a carbonate phase, though this could not be recognized optically. Verwoerd (oral communication) is at present carrying out a detailed study of the Zandkopsdrift pipe. He reports the presence of fenites in a stream bed at the foot of the hill, and suggests that the occurrence might prove to be a carbonatite complex.

A further striking feature of the Zandkopsdrift pipe is the

presence of irregular peripheral outcrops of massive manganese oxides. These contain minor amounts of the secondary uranium-thorium mineral betafite and also churchite - a Ca-rare earth-phosphate mineral (W J Verwoerd, oral communication).

Small irregular outcrops (up to 50 m across) of brecciated chert, cemented by manganese-rich oxides are closely associated with several, though apparently not all, of the other olivine melilitite pipes. In the Garies area the margins of such outcrops are often marked by the presence of numerous small aloes, whose restricted distribution suggests a dependence on the unusual chemistry of the surrounding soil. There seems little doubt that there is a genetic relationship between the olivine melilitites and associated manganese mineralization, although the significance of this association is still uncertain.

2.2 PSEUDO-KIMBERLITES

Exploration geologists in the Bushmanland region have identified a number of diatremes which have been termed "kimberlites" as typical kimberlitic indicator minerals (microilmenite, chrome diopside and purple Cr-rich garnets) are recovered from drill core concentrates. The surface expression of such "kimberlites" is generally a shallow depression with a thick calcrete crust. Isolated boulders of fresh olivine melilitite which are scattered about the surface of some of these "kimberlites" have been interpreted as xenoliths.

Samples KKV-1 and KKV-2 are respectively "kimberlite" and olivine melilitite that were recovered from the Klein Katvlei pipe.

The former was kindly donated by Mr H Jenner-Clarke. It is a grey-green highly altered specimen with a strongly pitted surface resulting from the selective etching of a phenocryst phase, probably olivine. The rock is broken into rounded fragments that are cemented by carbonates, which may be primary though this would be difficult to establish because of the abundance of associated calcrete. In overall appearance, the specimen closely resembles an altered and brecciated olivine melilitite. Analyses of KKV-1 and KKV-2 are listed in Tables 1(b) and 1(c) respectively. The most marked compositional differences are the higher SiO_2 and very much lower MgO contents of the "kimberlite" relative to the olivine melilitite. Concentrations of Zr, Nb, Y and P_2O_5 are closely similar in the two rocks, which is significant, as these elements are likely to be relatively unaffected by alteration. Concentration levels of most of the other components are broadly comparable. These similarities provide support for the suggestion that in the Namaqualand area, hydrothermally altered olivine melilitites (which in this work will be termed pseudo-kimberlites) may have been confused with kimberlites (Cornelissen and Verwoerd, 1975). However, whether or not all the Namaqualand "kimberlites" are merely altered olivine melilitites (i.e. pseudo-kimberlites) is difficult to establish at this stage, as the characteristically poor exposures make recovery of fresh material virtually impossible. Nevertheless, typical kimberlite indicator minerals have been recovered from pipes such as Klein Katvlei which probably are strongly altered olivine melilitites. It therefore seems that

a detailed study of the Namaqualand pseudo-kimberlites would be rewarding in terms of clarifying the genetic link between the olivine melilitites and true kimberlites.

2.3 SEDIMENT-FILLED (KIMBERLITE) PIPES AND BRECCIA NECKS

Reuning (1931) initially drew attention to the presence of sediment-filled diatremes to the south of Gamoep. He interpreted these pipes as being kimberlites. They have recently been described in greater detail by Cornelissen and Verwoerd (1975).

The pipes range in size from 50 m to 500 m in diameter and are filled by radially inward dipping lacustrine sediments to depths of up to 260 m. These sediments overlie blue ground that superficially resembles kimberlitic breccia. They include sandstone, grits, arkoses, conglomerates and shales. The latter may have intercollated tuffaceous material. Great blocks of country rock gneisses, apparently derived from a marginal wall rock breccia, have collapsed into the sediments close to the edge of some of the pipes. This sometimes results in rather spectacular soft-sediment deformation features (Plate 6).

A peculiar feature of the sediment-filled pipes is the presence of marginal late-stage opaline veins that are sometimes associated with thick manganese encrustations.

In the same general area as the sediment-filled pipes are a number of small breccia necks (Plate 5) composed almost entirely of a variety of angular gneissic rock fragments. These probably represent small craters that became choked by a collapsing

wall rock breccia.

Sediment-filled pipes have been found to the south of Garies (H Jenner-Clarke, oral communication), though are not as numerous as on the Bushmanland plateau. In general, the Garies pipes tend to have a shallower infill than the latter, probably because of the deeper level of erosion of the coastal Pliocene surface.

The excavation of deep palaeocraters and development of marginal wall rock breccias indicate violent eruptions. Explosive activity may have been triggered by either the large expansion of volcanic gases at low pressures, contact with ground water or a combination of these factors.

Sample Bit-1 (Table 1c) is the only specimen of blue ground from a sediment-filled pipe that has been analyzed during the course of this study. It is from a core section recovered from a pipe on the farm Mostertsvlei to the south of Garies, and was kindly donated by Mr H Jenner-Clarke.

2.4 AGE RELATIONSHIPS

Age relationships of the volcanic pipes and diatremes in the Namaqualand-Bushmanland area are still equivocal. Two zircon dates for the Gamoep kimberlites (Davis, 1977) indicate a late Cretaceous age (67 m.y.) for these volcanics. This is supported by fossil evidence (Haughton, 1931). There are however, no published dates for the associated olivine melilitites in this pipe cluster. A zircon recovered from the Mosterts-

olivine kimberlite to the south of Garies gives an age of 54.1 m.y. for this pipe (Davis, op. cit.). Kröner (1973) quotes an unpublished K/Ar age of 38 m.y. for an olivine melilitite pipe in the same area.

These data do not clearly indicate whether the olivine melilitites and kimberlites in the two pipe clusters represent separate episodes of igneous activity, or whether continuous volcanism took place during late Cretaceous and Tertiary times. However, the former possibility should not be discounted, and if the olivine melilitites in the two pipe clusters are of different ages, this may have bearing on the interpretation of bulk rock chemistry.

The significance of the Namaqualand-Bushmanland volcanics in relationship to post-Gondwanaland tectonics in southern Africa is discussed in Section 6.

3. PETROGRAPHY

Rogers (1911) and Taljaard (1936) have presented detailed petrographic descriptions of the olivine melilitites and related rocks from Namaqualand, while Gerrard (1958) describes those from elsewhere in the Cape Province. During the course of the present investigation, a large number of samples covering a broad compositional range (7.5% MgO - 24% MgO) were studied. This has made it possible to expand on earlier observations and further, by combining petrographic evidence and mineral chemistry data with recent advances in experimental petrology (e.g. Yoder, 1973; Donaldson, 1976; Mysen, et al., 1975 and others), to piece together a rather detailed model for the evolution of these magmas. The petrology of the Namaqualand volcanics is accordingly treated at some length. A general description is followed by more detailed descriptions of individual mineral phases.

3.1 GENERAL

The olivine melilitites are porphyritic rocks, with large olivines invariably present as a major and often dominant mineral phase. These commonly range in size from 2 mm in length down to groundmass microphenocrysts (Plate 7), indicating an extended period of crystallization. However, unusually large olivines up to 40 mm in length have been recovered from two pipes. It is possible to distinguish a number of different olivine groups as discussed ahead. Titanomagnetite is a major phase in all of the Namaqualand-Bushmanland pipes. In

some, though not all of these, this phase occurs as large anhedral (up to 10 mm in diameter). However, there is always a prominent population of titanomagnetite microphenocrysts, that show a continuous size range down to a groundmass dusting. Very rare microilmenite (with up to 11.6% MgO) has been identified in a number of pipes. When present, the mineral is almost invariably mantled by a rim of skeletal titanomagnetite fingers, which are in turn surrounded by an outer margin of perovskite grains (Plates 24 and 25). All of the Namaqualand-Bushmanland pipes are characterized by the presence of ubiquitous perovskite microphenocrysts. These typically occur as equant anhedral, rude cubes, or as characteristic interpenetrant twins. Melilite microphenocrysts are abundant in some specimens, but absent in others. Apart from the absence of this phase, melilite-free rocks are mineralogically similar to the associated olivine melilitites, and will be termed olivine nephelinites to distinguish them from the latter. Factors governing the presence or absence of melilite are considered ahead (Section 3.2).

There is a range in coarseness of the groundmass assemblage of the olivine melilitites and olivine nephelinites. In coarse-grained specimens, the groundmass is composed of an interlocking mosaic of clinopyroxene laths (up to 0.5 mm in length), with interstitial nepheline, a sprinkling of titanomagnetite and a second generation of granular perovskite. Apatite has been identified microscopically in the coarsest specimens. A sulphide phase, identified as pyrite in reflected light, is usually finely disseminated as a late-stage interstitial mineral, but varies considerably in modal proportions from one

specimen to another. Phlogopite is sometimes present as a late-crystallizing phase in coarse-grained specimens, and may marginally replace earlier olivine phenocrysts. The phlogopite generally shows normal pleochroism, but in the Hoedkop pipe (code-named Ho) both normal and reversed pleochroic varieties are present. This is of interest as reversed pleochroism in phlogopite is common in kimberlites.

In finer-grained specimens, the groundmass is dominated by tiny (0.02 mm) clinopyroxene needles together with a second generation of granular perovskite, a dusting of titanomagnetite and a finely disseminated sulphide phase, forming a dense mat. Microprobe x-ray images of fine-grained specimens show the presence of tiny grains of a Ca- and P-rich phase, probably apatite, which is thus considered to be a common late-crystallizing mineral. Glass and nepheline are probably also present, though can seldom be positively identified. Phlogopite is not common in fine-grained varieties.

Modal mineral proportions for selected samples covering a range in bulk rock chemistry are given in Tables 5(a) and 5(b).

3.2 DETAILED PETROGRAPHY AND MINERAL CHEMISTRY OF THE DOMINANT MINERAL PHASES

The different mineral phases show a variety of textural relationships that have important bearing on the origin of the olivine melilitites, and these are now discussed in more detail.

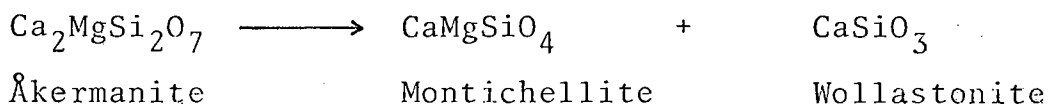
3.2.1 MELILITE

3.2.1.1 *General*

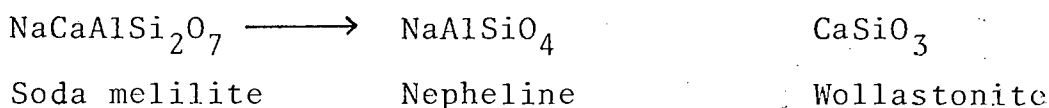
In the Namaqualand volcanics, melilite is present in samples covering a wide compositional range (9.9% MgO to 20.2% MgO). However, within this chemical spectrum, there are melilite-free varieties (olivine nephelinites). The occurrence of melilite is sometimes regarded as being important in establishing the kinship of alkaline ultramafic volcanics. Thus, petrologists from De Beers Consolidated Mines regard the presence or absence of this mineral as an important criterion for distinguishing non-kimberlitic rocks from true kimberlites (Roger Clement, oral communication). It therefore becomes important to understand the factors that govern the presence of the phase.

Textural relationships indicate that the occurrence of melilite in the Namaqualand volcanics was to a large degree controlled by the emplacement and cooling history of the different magmas. In fine-grained rocks, the mineral occurs as tiny laths (0.1 mm in length) that may show a patchy distribution and flow alignment about earlier olivine phenocrysts (Plate 23). However, in specimens with a coarser groundmass assemblage, melilite typically occurs as large evenly distributed plates, up to 0.4 mm in length, that are often marginally replaced by clinopyroxene, producing a characteristic hourglass shape (Plate 22) while, within the spread of melilite-bearing compositions, those specimens with an extremely coarse groundmass lack melilite. It is suggested that these

petrographic features can be explained in terms of crystallization of the mineral over a limited depth range followed by a sub-solidus breakdown during slow cooling at low temperatures and pressures by analogy with the reactions:



and



which have been documented by Yoder (1973). The wollastonite would be represented by groundmass clinopyroxene in the natural system, and the montichellite by the marginal Ca-enrichment that is characteristic of the olivines (as described ahead).

3.2.1.2 *Experimental Evidence*

Experimental evidence summarized by Yoder (op. cit.) indicates that melilites with compositions that are characteristic of volcanic rocks are unstable at elevated pressures. Pure $\text{\AA}kermanite$ ($\text{Ca}_2\text{MgSiO}_7$) is stable to 14 Kb under anhydrous conditions (Fig. 3.1) and stability is lowered to 10.2 Kb in the presence of excess H_2O (Fig. 3.2) and 8.7 Kb in the presence of excess CO_2 (presumably dissolved in silicate liquids as a carbonate) (Fig. 3.3). Yoder (1973) notes that common igneous melilites have compositions close to $\frac{1}{3}$ soda melilite: $\frac{2}{3}$ $\text{\AA}kermanite$, with only minor solid solution towards gehlenite ($\text{Ca}_2\text{Al}_2\text{SiO}_7$). This would imply stability up to pressures

of 15-20 Kb under anhydrous conditions at 1 000°C (Fig. 3.5). The effects of H₂O and CO₂ have not been documented for such compositions, but might reasonably be expected to lower the stability range to pressures below about 10-15 Kb. Melilite crystallization in natural igneous systems rich in CO₂ and H₂O would therefore only be expected at depths shallower than 30-45 km. Rapid magma ascent over this depth range followed by rapid chilling probably accounts for the smaller melilite microphenocrysts of the fine-grained rocks, while slower ascent, or slower cooling on emplacement would permit growth of the larger laths characteristic of the coarser specimens. Extremely slow cooling rates would result in the elimination of the phase through sub-solidus breakdown to clinopyroxene, by analogy with the reaction presented above. In addition to these factors, the stability of melilite is probably affected by the alkali content of the magma during crystallization (Velde and Yoder, 1976). If these conclusions are valid, the presence or absence of melilite cannot be regarded as a reliable criterion for determining the kinship of olivine melilitites and other alkaline ultrabasic magmas - particularly those such as kimberlites that are rich in volatiles like CO₂ that have a marked effect on melilite stability.

Melilite is absent from the most MgO-rich volcanic rocks from Namaqualand (samples ZHM-3, ZHM-4 and ZHM-5). As these are all very coarse-grained, the absence of the phase may once again be related to magma cooling histories. However, it is possible that chemical factors such as the relatively low Ca

concentrations in these rocks may also be important, although these cannot be evaluated from available data.

3.2.2 OLIVINES

3.2.2.1 Petrography

Petrographic criteria make it possible to distinguish several olivine groups, although it will be shown that these constitute a chemically related sequence and that the boundaries between the different populations, while useful for descriptive purposes, are rather artificial. Not all of the groups have been recognized in every pipe, but it is hoped to demonstrate that the petrographic descriptions, in combination with chemical data that are to follow, are generally applicable to all of the Namaqualand olivine melilitites.

- (a) Hopper olivines. The dominant type of olivine in all pipes (invariably more than 95% of all olivines) are euhedral and sometimes skeletal olivines that will be termed "hoppers" as they bear a striking resemblance to the growth forms described by Donaldson (1976), with re-entrants and internal cavities. They have sharply defined crystal edges and corners in contrast to the rounding that would be expected from resorption processes. Representatives of this population are illustrated in Plates 7, 8, 10 and 11. Hopper morphology is best represented in the Mg-poor volcanics, possibly reflecting compositional control on growth forms (Donaldson, op. cit.).

The hopper olivines enclose a variety of different types of inclusions:

- (i) Most common are what will be termed "composite inclusions" (Plates 18 and 19) as they generally comprise a number of different phases: zeolites, chiefly natrolite and thompsonite, are usually the most abundant of these, though one or more of the minerals clinopyroxene, titanomagnetite, phlogopite, perovskite and pyrite may be present in varying proportions. The composite inclusions are often markedly coarser grained than the groundmass assemblage in the same rock. They are considered to have crystallized from liquids trapped during olivine growth (Rogers, 1911). The abundant zeolites in these inclusions indicate that the trapped liquid was extremely hydrous, suggesting affinities with the immiscible pegmatite liquids described in Section 3.3, rather than the drier olivine melilitite liquids.
- (ii) Very rare rounded or pear-shaped carbonate inclusions, up to 0.1 mm in diameter, have been identified in hopper olivines in samples from a number of different pipes (Plate 20). Semi-quantitative microprobe data show the carbonate to be Ca- and Sr-rich (up to 9400 ppm Sr) and Mg-poor. The characteristic shape of these inclusions indicates that they were trapped as a

liquid rather than a solid, which suggests that an immiscible carbonatitic fluid was associated with the olivine melilitites at the time of hopper olivine crystallization. If so, this fluid must have been lost during the evolution of the olivine melilitite magmas, for carbonates are not primary crystallizing phases in these volcanics. The small carbonate dyke associated with the Zandkopsdrift pipe (Section 2) may represent such an immiscible carbonate liquid.

- (iii) Round or oval sulphides form a third distinct variety of inclusions in hopper olivines, though are extremely rare. Their morphology suggests that they were also trapped as liquids rather than solids and therefore that an immiscible sulphide liquid may have been present during the period of hopper olivine crystallization.
- (iv) Minute fluid inclusions, some with bubbles, are present in many of the hopper olivines. Isolated individuals are interpreted as being primary, and these point to the existence of a free vapour phase during the period of hopper olivine crystallization. Inclusions which form randomly oriented trains that traverse the olivine interior are probably secondary and mark the site of annealed fractures. A study of fluid inclusions in the Namaqualand volcanics would undoubtedly

be highly rewarding, but is beyond the scope of the present investigation.

- (b) HILN olivines. Representatives of another olivine population will be termed HILN olivines because of their characteristic chemistry, which is discussed ahead. Olivine 5 in Plate 7 and the olivines illustrated in Plates 14 and 15 are examples of this olivine population. They can be distinguished petrographically by their relatively large size, the absence of marked undulose extinction, evidence of strong marginal resorption and by the presence of yellow glassy blebs that are considered to indicate partial melting of the olivine. Such glassy blebs are often concentrated about the olivine margins. These olivines are typically anhedral or subhedral (Plate 14) although occasionally show marginal remnants of an originally euhedral form (Plate 15). Trains of fluid inclusions that traverse the olivine interiors are abundant. These presumably represent annealed fractures. Rare rounded carbonate and composite inclusions similar to those described in the hopper olivines are also found. One of the composite inclusions differs from those typical of the hopper olivines in containing a euhedral and apparently primary calcite rhomb in addition to the dominant zeolites and clinopyroxenes (Plate 21).
- (c) Megacryst olivines. A further group of olivines comprises exceptionally large, coarsely fractured indivi-

duals, up to 40 mm in greatest dimension (Plates 16 and 17). These are generally rounded, although some have relatively straight edges that might be rational crystal faces. Narrow zones of mosaic texture in these olivines indicate incipient recrystallization although large segments of the crystals are usually relatively strain-free. Following the usage in kimberlite studies (e.g. Nixon and Boyd, 1973; Gurney et al., 1977), the non-genetic term megacryst will be applied to these unusually large olivines. Any coarsely fractured, rounded olivine greater than 10 mm in diameter may confidently be included in this population, but the choice of a lower size limit for the megacrysts is quite arbitrary, as disaggregated remnants are likely to be present in megacryst-bearing pipes. Megacrysts are only present in any quantity in two pipes from the northern cluster, and have not been found in any of the southern cluster of pipes. The megacryst-bearing pipes are unusual in having numerous ultrabasic xenoliths of upper mantle derivation. Spinel harzburgites are the most common of these exotic inclusions, but rare garnet lherzolites have been found in one of the pipes (Moore, 1973). The reason for the apparent restriction of the megacrysts and ultrabasic xenoliths to certain pipes is not clear, but may be related to the interplay of magma ascent velocities and settling rates of the inclusions (Kushiro et al., 1976).

(d) Xenocryst olivines. There is, in addition, a group of rare strained anhedral grains that can be distinguished petrographically from the second population by their undulose extinction and lack of evidence of incipient melting. These olivines are only found in those northern pipes with abundant ultrabasic xenoliths.

The question of whether the different olivine populations are phenocrysts or not is very much the substance of this thesis. It is hoped to demonstrate that the first three groups are cognate, and can be related in terms of a continuous magma evolution model, and that the olivines of the fourth group are probably xenocrysts.

3.2.2.2 *Olivine Chemistry*

Analytical techniques. Analyses were carried out on a Cambridge Microscan-5 electron microprobe at 20 kv and 1500 uA and data were reduced using Bence-Albee correction factors. The Ni standard was an olivine (0.29% Ni) from a dunite which had previously been analysed by x-ray fluorescence and subsequently checked against a pure Ni metal standard. Ni analyses generally entailed 100 second counts on the K_{α} peak, and 50 second background counts on either side of the peak, giving a lower limit of detection of 120 ppm Ni (99% confidence limit) and one standard deviation of 40 ppm based on the counting statistics of the standard.

Results. Olivines (excluding the rare megacrysts) have been analysed in nine rock specimens from four pipes in the south-

ern cluster and four from the Gamoep group. The data indicate that the most significant chemical changes are shown by Mg, Fe, Ni and Mn, and that compositional variation in these elements is extremely complex in olivines from the same rock. A particularly detailed study was made of the olivines in several thin sections cut from one specimen from the Dikdoorn pipe in the southern cluster, namely Dik-9; bulk rock Mg/(Mg + Fe) (atomic proportions) = 0.563; Ni (weight percent) = 319 ppm.

In addition, analyses were made of chips from 14 olivine megacrysts recovered from the Zwartheuwel pipe, which forms a low prominent hill one kilometre to the north of Gamoep. Spot analyses were carried out on a thin section cut from one of the larger megacrysts (Plate 17) in order to establish whether chemical zoning was present.

Chemical criteria make it possible to recognize several distinct olivine groups, which can be correlated with those recognized on petrographic grounds. These will first be discussed with reference to the olivines from Dik-9 (Figs. 3.6(a) and 3.6(b)).

- (a) Hopper olivines. The interiors of most hopper olivines collectively define a compositional trend of decreasing Mg/Fe with gently decreasing Ni, while the hopper margins are characterized by a narrow rind (up to 300 μ m) that shows continued Ni depletion, but strong reversed zoning with respect to Mg/Fe (see Figs. 3.6(a) and 3.6.(b) together with marked

Ca-enrichment (sometimes over 1% CaO). The margin compositions correspond to those of late-crystallizing olivine microphenocrysts, and apparently reflect partial equilibration of the hopper in response to late-stage changes in the magma composition or conditions of crystallization. These zonation patterns are mirrored by individual hopper olivines. Olivines 8, 9, 11, 15, 23, 24, 37, 39, 42 and 52, which are illustrated in Plate 7, are examples of individuals which show such behaviour.

In contrast to the zoning trends described above, a number of hopper-form olivines show aberrant zonation patterns. These individuals will be referred to as "unusual" hoppers, for they are commonly euhedral, may have inclusions similar to those found in the hoppers but are not petrographically distinctive, apart from the fact that some are relatively large, and not markedly skeletal. Examples are olivines 1, 6 and 13 in Plate 7. There are, however, large hoppers which do not show abnormal chemical variations. The centres of the "unusual" hoppers differ considerably in terms of both Mg/(Mg + Fe) and Ni content from "normal" hoppers and there does not appear to be any rational pattern to these variations. The margins of all the "unusual" hoppers are characterized by chemical zonation towards "normal" hopper centres or groundmass compositions. Depending on the core composition, this may result in either normal (e.g. Dik-9 olivine 6) or reversed (e.g. Dik-9-3 olivine 7) zoning with respect to Mg/Fe, and

generally leads to a narrow zone of Ni enrichment close to the olivine margin followed by an outer rind characterized by Ni-depletion but Mg-enrichment, reflecting zonation towards groundmass compositions (Figs. 3.6(b) and 3.7).

(b) HILN olivines. The anhedral or subhedral olivines of the second population are enriched in Fe and Mn (up to 1.5% MnO in some individuals) and markedly depleted in Ni relative to the hopper olivines in the same rock specimen. These high iron, low nickel (HILN) olivines show marginal zonation analogous to that displayed by the hoppers, resulting in HILN edges being enriched in Mg and Ni relative to the core (note that the hoppers are relatively enriched in Mg, but depleted in Ni at the edges). The data indicate that the HILN olivines show a relatively large spread in $Mg/(Mg + Fe)$ in all rocks studied.

$Mg/(Mg + Fe)$ vs. Ni data for olivines in two further specimens - Bies-3 from the southern and WK-3 from the northern pipe cluster - are shown in Figs. 3.9 and 3.10. The olivine varieties recognized in Dik-9 (hoppers, "unusual" hoppers and HILN olivines) are present in both rocks and show analogous chemical variations to those described for Dik-9 olivines. Fig. 3.10 does not clearly indicate the marginal reversed zoning of WK-1 hoppers. This is, however, a reflection of the limited number of data points, for Mg and Fe step scans across individual olivines show that such reversed zoning is present.

It is therefore believed that many of the Namaqualand melilitites are characterized by having diverse olivine populations analogous to those recognized in Dik-9. Significantly, this appears to be true for both the northern and southern pipe clusters.

Other pipes. A small number of olivines have been analysed in several other rock specimens (Figs. 3.11 to 3.19). As in Dik-9, there is always a range in composition of olivines from the same sample, although possibly because of the limited number of data, chemical trends are not well defined in all cases. However, euhedral olivines in sample ZH-14 show a well-defined zonation pattern (Fig. 3.19) that is markedly different from that found in Dik-9. Discussion of this zonation pattern is reserved until a later stage (Section 3.2.2.4(g)) although it will be demonstrated that it can be understood in terms of the general model proposed to explain the variation of olivine chemistry in the Namaqualand volcanics.

(c) Megacrysts. Mg/(Mg + Fe) - Ni relationships defined by the Zwartheuwel megacrysts are illustrated in Fig. 3.18. (As noted previously, olivine megacrysts have not been found in any of the pipes in the southern cluster.) The olivine megacrysts analysed collectively define a fairly regular trend of decreasing Ni and increasing Mn towards fayalitic compositions (Fo_{92.2}, 0.36% Ni, 0.07% Mn to Fo_{74.7}, 0.17% Ni, 0.26% MnO). This overall pattern of variation is similar to that

found for megacrysts from the Monastery kimberlite (Gurney et al., 1977). The range in composition of chips from the same megacryst (Fig. 3.18) suggests that these olivines are strongly zoned. However, spot analyses across a thin section cut from one of the megacrysts (ZH-14) show this olivine to be chemically homogeneous except where strongly altered. The sense of chemical variation in such altered zones indicates re-equilibration towards groundmass compositions, and this suggests that the megacryst was initially homogeneous. It is significant that the most fayalitic of the Zwartheuwel megacrysts are depleted in both Mg and Ni relative to the euhedral olivines from the same pipe.

- (d) Xenocrysts. The strained anhedral olivine population are unzoned and have compositions (Fe_{91} , 0.40% NiO) that fall within the range found for presumed mantle olivines (Reid et al., 1975). The chemistry, morphology and limited occurrence of such individuals suggests that they are either xenocrysts derived from disaggregated mantle material, or fragments of Mg-rich megacrysts. They are therefore excluded from further discussion. Like the megacrysts, such olivines have not been found in the Dikdoorn pipe.

In summary, it appears possible to recognize the following olivine groups in many of the Namaqualand pipes:

- (a) Euhedral (hopper) olivines, most of which have cores

that show normal zoning with respect to Mg/Fe and Ni, but often have narrow rims characterized by strong Mg-enrichment. A number however, show aberrant zonation patterns ("unusual" hoppers).

- (b) Anhedral or subhedral (HILN) olivines that show evidence of incipient melting and which are characterized chemically by high iron and manganese and extremely low Ni concentrations relative to the hoppers from the same rock.

In addition, some of the northern pipes have megacrysts, up to 40 mm in diameter. These olivines are homogeneous except in strongly altered areas, which show zonation towards groundmass olivine compositions. The megacrysts collectively show a wide spread in MgO and Ni. The most fayalitic specimens are depleted in Mg and Ni relative to small euhedral olivines from the same pipe. The reason for the absence of megacrysts in many of the pipes is uncertain, though may be largely related to the interaction of magma ascent rates and settling velocities.

A final group of anhedral olivines show strained extinction and are relatively enriched in Mg and Ni. These are only common in those pipes with abundant ultramafic xenoliths.

Any model for the origin of the olivine melilitites should explain the wide range in olivine compositions in individual pipes, and the complex zonation patterns found in the "unusual" hoppers. This however raises one of the fundamental problems

confronting the igneous petrologist - that of distinguishing between cognate and exotic phases. The characteristic euhedral form of the "normal" hoppers leaves little doubt as to their being cognate phenocrysts. The strained anhedral of the fourth petrographic olivine population are probably either exotic or fragments of megacrysts. It is therefore necessary to consider whether the "unusual" hoppers, HILNS and megacrysts are cognate or accidental.

3.2.2.3 *Origin of Olivines*

The well-developed crystal outline of the "unusual" hopper olivines (Plates 12 and 13) suggests that they are cognate phenocrysts. However, Ni and Mg step scans across a number of these individuals define narrow zones, usually close to the margins, which display marked compositional gradients (Fig. 3.7). It could, therefore, be argued that the cores of these olivines are exotic and that euhedral outline is an artifact of overgrowth subsequent to incorporation in the olivine melilitite magma. Although this possibility cannot be dismissed, it should be noted that the strained olivine anhedral which are considered to be exotic invariably show marginal resorption features rather than overgrowths. In addition, some of the "unusual" hoppers contain composite inclusions similar to those that characterize the "normal" hoppers, which indicates that the liquids trapped by these olivines were broadly similar.

Two HILN olivines with well-preserved remnants of an original

euohedral morphology have been recognized (e.g. Plate 15), and there are several which could be described as subhedral. These olivines typically show varying degrees of marginal re= sorption, and the presence of rational crystal edges in some individuals therefore seems more likely to indicate an origi= nally euohedral morphology than to result from overgrowths to exotic xenocrysts. Moreover, the unusual compositions of HILN olivines are atypical of those found in presumed mantle= derived olivines, or those commonly reported from crustal rocks, and appear to be unique to the olivine melilitites, which strongly suggests a cognate origin. Further, HILN oli= vines are never found intergrown with a second phase, as might be expected if they were accidental. As we have iden= tified HILN olivines from both the northern and southern pipe clusters - a lateral extent of some 150 km - any postulated exotic source rock would probably have to be areally extensi= ve. An additional, and compelling line of evidence for a cognate origin for the HILN olivines is the broad mineralogi= cal similarity of the composite inclusions in these and hop= per olivines, which suggest crystallization from similar mag= mas. However, the presence of an apparently primary carbona= te rhomb in one of the HILN composite inclusions indicates that the liquid from which these olivines crystallized may have contained relatively more dissolved carbonate than that which precipitated hopper olivines.

The status of the megacrysts is not as well defined as that of the other olivine groups, but several lines of reasoning

suggest that they are also cognate phenocrysts. The fairly regular chemical variation shown by this population indicates that they are genetically related. The morphology and chemical characteristics of these olivines indicate a close affinity to the olivine megacryst suites that have been recovered from several kimberlites (Nixon and Boyd, 1973; Gurney et al., 1977). The latter authors strongly favour a cognate origin for the megacrysts. Gurney (oral communication) points out that the compositional ranges, and the absence of strain features in kimberlite megacrysts, are very atypical of olivines found in ultrabasic xenoliths of apparent upper mantle origin. There is a further indirect reason for regarding the megacrysts as cognate. O'Hara (1965) points out that the first phases to crystallize from magmas of mantle derivation would normally be the same minerals (with identical compositions), as those which formed the residue in the source area. Olivine is almost certainly a residual mineral during most mantle melting events, and because the olivine primary phase field expands towards lower pressures (O'Hara, 1968), the mineral would be expected to be an early crystallizing phase in mantle-derived magmas. If so, early-formed olivines should match those in the source rocks, and would be expected to range in composition from Fo₈₈ to Fo₉₃. It is suggested that the most Mg-rich megacrysts (Fo₉₂) might represent such early crystallizing olivines. Olivine megacrysts have not been found in all the pipes - for example, the Dikdoorn pipe. Nevertheless, as it is believed that their presence or absence might reflect the balance between

settling rates and magma ascent velocities, it is doubtful whether the absence of olivine megacrysts implies non-crystallization. Rather it seems likely that such olivines may have crystallized from all of the Namaqualand olivine melilitite magmas.

It is therefore believed that the megacrysts, HILN and "unusual" hopper olivine populations are cognate. Certainly to dismiss them as accidental inclusions only drives the problem of their origin one stage further back. It is admitted that the chemical trends shown by the rims of these olivine populations does allow for an exotic origin, but for the reasons given this possibility is not favoured. Magma mixing processes offer a potential explanation for the heterogeneous olivine populations that characterize the Namaqualand olivine melilitites. However, although 58 bulk rock samples from 21 pipes have been analysed, no evidence has been found for the existence of an Fe-rich and Ni-poor liquid that might have crystallized HILN olivines. Moreover, extremely complex magma mixing processes, presumably involving a variety of different liquid compositions, would be required to account for the spread in Mg/Fe and Ni found for the cores of the "unusual" hoppers. A magma mixing model to account for olivine compositional complexity is thus not favoured. Presented below is a model which assumes that all the olivine populations, (with the possible exception of the strained anhedral of presumed mantle composition) are phenocrysts which reflect chemical and physical variations related to the evolution of the host

magmas. If valid, this model has important implications to the genesis of kimberlites and diamonds.

3.2.2.4 Discussion

- (a) Oxygen activity control and HILN compositions. It is convenient to reserve discussion of the megacrysts until a later stage, for the origin of these olivines follows naturally from the relationship between HILN and hopper olivines.

On the basis of their morphology (i.e. evidence of resorption and melting in contrast to the sharply defined edges of the hopper olivines) and their limited abundance, it seems reasonable to assume that HILNS crystallized before the hopper olivines, and further, that they crystallized from a magma which did not have unusually low Mg and Ni contents (in view of the hopper compositions). It is also unlikely that the HILN compositions result from low Ni^{++} distribution coefficients since the high temperatures (Leeman, 1974) or high MgO contents (Hart and Davis, 1978) necessary to achieve this would result in the crystallization of a more magnesian olivine. It is therefore suggested that the compositional characteristics of the HILN olivines in part reflects crystallization at low oxygen activities, significantly below the Ni/NiO buffer, but above the Fe/FeO buffer (see Fig. 3.8 for the relative positions of different buffer curves). Low oxygen activity would result in high Fe^{2+}/Fe^{3+} , Mn^{2+}/Mn^{3+} and Ni^0/Ni^{2+} ratios

in the magma, thus producing high Fe and Mn combined with low Ni in the olivines crystallizing in equilibrium with such a liquid.

The crystallization of hopper olivines could have taken place at higher oxygen activity (above the Ni/NiO buffer), giving rise to substantially higher Mg/Fe ratios and higher Ni contents in the olivines crystallizing under these conditions. The Mg and Ni-enriched rims surrounding the HILN olivines may reflect partial re-equilibration under higher oxygen activity conditions, or may be overgrowths under these conditions. The trend of decreasing Mg/Fe ratio and decreasing Ni content from the interiors to the margins of the hopper olivines is seen as a response to falling temperature and the progressive removal of Ni by Raleigh-type crystallization. The reversed-zoned hopper margins may reflect an increase in the Mg/Fe⁺⁺ ratio of the magma caused by extensive crystallization of Fe-Ti oxide phases or extreme late-stage oxidation of the magma, or both. Titanomagnetite microphenocrysts are often partially enclosed by olivines in rocks from the southern cluster (though not in those from the northern pipes). This petrographic evidence indicates that crystallization of the opaque phase coincided with the late-stage of olivine growth in some of the magmas.

Having suggested major changes in oxygen activity during the crystallization of the olivine melilitite magmas, it

is necessary to consider a possible explanation for such changes.

The presence of primary carbonate in one of the HILN composite inclusions suggests that the coexisting equilibrium liquid may have been relatively carbonate-rich compared to bulk rock compositions of the olivine melilitites, which typically contain less than 0.3% CO₂ (Table 1(1)). Recent experimental studies, for instance Brey and Green (1977), have stressed the importance of CO₂ in the generation of olivine melilitite magmas. The CO₂ would in part be dissolved as a carbonate in the primitive melt according to these authors. It seems plausible that a gas buffer involving CO₂ may have maintained the olivine melilitite magmas at low oxygen activities, possibly close to or on the carbon saturation surface, during part of the evolutionary history of these magmas. The relative positions of relevant buffer curves are shown in Fig. 3.8. Equations summarized by Heubner (1971) indicate that the carbon-gas buffer will be below the wustite-magnetite buffer, and thus well below the Ni/NiO buffer at high temperatures and pressures (1 000 - 1 300°C; 10-15 Kb). Under such conditions, $a_{\text{Ni}^{2+}}$ in the liquid will be held at low values and reduced Ni may be formed, while $\text{Fe}^{3+}/\text{Fe}^{2+}$ and hence Mg/Fe^{2+} ratios will be relatively low. These conditions of low oxygen activity are considered to be in part responsible for the compositional characteristics of the HILN olivines. The high Mn content of the HILN olivines is

considered to be related partly to high Mn^{2+}/Mn^{3+} in the highly reduced magma, though may in addition indicate that Mn is strongly partitioned into olivine in CO_2 -rich magmas (with CO_2 dissolved as a CO_3^{2-} anion).

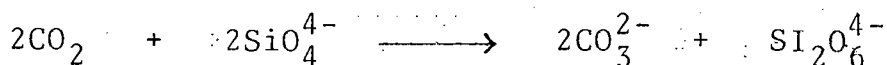
It is proposed that reduction in pressure during magma ascent would initiate the loss of volatiles and dissolved carbonate, which in turn would lead to a substantial increase in oxygen activity and hence in Ni^{2+}/Ni^0 , Fe^{3+}/Fe^{2+} and Mg/Fe^{2+} ratios in the melt. (The reason for such an increase in oxygen activity is discussed in Section 3.2.2.4(e).) Furthermore, the loss of volatiles would cause an increase in liquidus temperatures, and this could result in rapid crystallization of relatively Mg- and Ni-enriched hopper olivines and also incipient melting of the earlier HILN olivines. The carbonate and fluid inclusions in hopper olivines are believed to record the loss of volatile components. The typically rounded morphology of the carbonate inclusions suggests loss of this phase as an immiscible liquid.

- (b) Variation in distribution coefficients and HILN olivine compositions. A consideration of the relationship between bulk rock chemistry and HILN olivine compositions shows that, even for the most favourable case where all iron is present as Fe^{2+} , the distribution coefficient of 0.3 suggested by Roeder and Emslie (1970) would only allow the most Mg-rich HILN olivines to be in equilibrium with the host bulk rock compositions, if the latter

are assumed to approximate liquid compositions. It will be argued on the basis of bulk rock chemistry (Section 4) that this assumption is plausible. However, even if the more magnesian specimens were enriched in cumulus olivine (and hence in Mg), the same seems unlikely to hold for relatively Mg-poor compositions, for example the rock Dik-9, which also contains HILN olivines that are markedly more Fe-rich than would be predicted by Roeder and Emslie (op. cit.). Similarly, while low oxygen activity would in principle favour crystallization of a more manganese-rich olivine from any given magma, it seems unlikely that such a control is alone sufficient to account for the gross difference in MnO between HILN olivines (up to 1.50% MnO) and hoppers (generally 0.15 to 0.3% MnO) as the Mn^{2+}/Mn^{3+} ratio is likely to be high even in comparatively oxidized magmas because of the relatively high oxygen activities defined by the $MnO-Mn_3O_4$ buffer (Heubner and Sato, 1970). To account for such apparent inconsistencies, it is suggested that transition element partition coefficients determined for olivine and basaltic liquids are inapplicable to carbonate-rich liquids such as those envisaged as the primitive olivine melilitite protomagmas. Two mechanisms, which may be complementary, could account for the differences in olivine-liquid partitioning for the carbonate-rich and carbonate-free systems.

(i) It has been suggested (Fraser, 1975, 1977) that

when CO_2 dissolves in silicate magmas, there is a change in melt structure as a result of the following polymerization reaction:



Burns (1969) has interpreted enrichment of transition elements such as Fe and Ni in olivine relative to coexisting orthopyroxene (for example in the suite of accidental peridotites derived from kimberlites), in terms of the higher crystal field stabilization energy (CFSE) for these elements in the octahedral sites in the unpolymerized olivine structure relative to those of the more polymerized pyroxene structure. If such an interpretation is valid, it might be expected that a change in melt structure would affect the site preference of several transition elements in the magma relative to coexisting olivines. By analogy with pyroxene-olivine partitioning, it is likely that an increase in the degree of polymerization of the melt would cause changes in partition coefficients that result in crystallization of olivines enriched in Fe, Mn and Ni relative to those in equilibrium with a less polymerized melt, other factors being equal. In other words, as the melt structure became more polymerized, the values of $K_{\text{Ni}}^{\text{ol-melt}}$, $K_{\text{Mn}}^{\text{ol-melt}}$ and the Mg/Fe distribution coefficient (K_D) as defined by Roeder and Emslie (1970), would all increase.

This interpretation is closely analogous to that of Watson (1976) and Hart and Davis (1978) who conclude that partitioning of the transition elements is strongly controlled by the Si-O ratio of the melt. In the case of Ni partitioning in the olivine melilitite protomagmas, the effect of a higher partition coefficient would be offset by low oxygen activities which would, in turn, lead to low $a_{\text{Ni}^{++}}$ values. (Note however, that provided such low oxygen activities do not fall below the Fe-FeO buffer, high values of $a_{\text{Mn}^{++}}$ and $a_{\text{Fe}^{++}}$ will be favoured in the magma.)

- (ii) In a carbonate-rich magma, it is possible that ion-pairing between Mg^{++} and CO_3^{2-} ions may result in a lowering of $a_{\text{Mg}^{++}}$ in the magma, and hence in a relative Fe-enrichment in the coexisting olivine. However, such a process seems inadequate to account for the marked Mn enrichment in HILN olivines relative to the hoppers, and would probably be secondary to variations in partition coefficients resulting from changes to the melt structure.

In summary therefore, it is suggested that the HILN olivines crystallized under conditions of low oxygen activity from a carbonate-rich magma. In addition, olivine-melt partition coefficients for the transition elements were higher than those appropriate to carbonate-free magmas of equivalent composition. The effects

of low f_{O_2} and high partition coefficients would be complementary with respect to Fe and Mn partitioning, but in the case of Ni, the increase in olivine-melt partition coefficient would be negated by the effects of low oxygen activity. As a result, the geochemical behaviour of Ni would be decoupled from that of the remaining transition elements, despite a relatively high CFSE in octahedral sites.

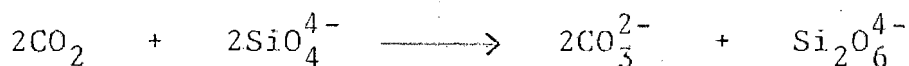
- (c) "Unusual" hopper compositions. The foregoing considerations suggest a possible explanation for the apparently irrational spread in Mg/Fe and Ni that characterizes the cores of the "unusual" hopper olivines. It is proposed that such chemical variability reflects crystallization during the early stages of loss of volatiles and an immiscible carbonate fraction from the magma as a result of large relative changes in pressure associated with rapid magma ascent at shallow levels (30-60 km to the surface). Loss of such components would be expected to result in independent changes to oxygen activity and olivine-melt partition coefficients for the transition elements, and the interaction of these effects would determine the composition of the cores of the "unusual" hoppers, which would then zone towards the compositions of the later crystallizing, more "normal" hoppers. The fact that the Ni-enrichment "peak" and Mg depletion "trough" do not coincide in "unusual" hopper 6 from Dik-9 (Fig. 3.7) is consistent with the variation

of the two elements being controlled by different processes.

- (d) Megacryst compositions. The megacrysts are interpreted as recording changes affecting the magma subsequent to separation from the source area, but prior to crystallization of HILN olivines. The most Mg-rich megacrysts (Fo_{92} , 0.36% NiO) are believed to be similar in composition to olivines in the source area. It seems unlikely that the spread in megacryst compositions reflect decreasing Mg/Fe and Ni in the host magma resulting from progressive Raleigh-type fractionation of olivines, for the most fayalitic megacrysts are depleted in Mg and Ni relative to the later crystallizing hopper olivine cores. Yet their large size, rounding and chemical range shows that they must undoubtedly have crystallized prior to the hopper population, granted the postulate that they are phenocrysts. The Mg-enrichment in hopper olivines relative to the megacrysts in the Namaqualand volcanics could in principle be ascribed to extensive ilmenite fractionation following megacryst crystallization. This would increase the Mg/Fe ratio of the residual magma, and hence that of the coexisting hopper olivines. However, such a process would also lead to a rapid depletion of the magma in Ti and Fe, and seems unlikely in view of the characteristically high concentrations of these elements in the olivine melilitites (4.2% to 7.6% TiO_2 ; 13.5% to 22.9% total iron as Fe_2O_3). In

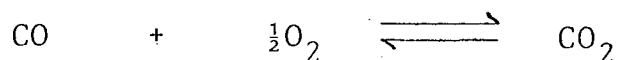
addition there is no petrographic evidence for extensive early crystallization of Fe-Ti phases. Similar objections apply to interpreting the increase in Ni concentration in hopper olivines relative to megacrysts in terms of ilmenite fractionation. Extensive fractionation of this phase would be needed to achieve the required increase in Ni concentration in the residual liquid and hence in the hopper olivines, even assuming that $K_{Ni}^{\text{ilmenite-liquid}}$ were appreciably less than 1. It is therefore suggested that the range in megacryst compositions is dominantly controlled by changing olivine-liquid partition coefficients during magma evolution as discussed below.

- (e) Physico-chemical considerations. Mysen et al. (1975) present evidence which suggests that at high pressures, CO_2 is present in silicate melts both as a carbonate and as a dissolved gas. Mysen (1976) reports that in albite-rich melts, the molar proportion of carbonate (defined as $X_{CO_3}^m = CO_3^{2-} / (CO_3^{2-} + CO_2)$) increases with decreasing pressure. He implies that the same is likely to be true for all silicate melts. Thus, a magma which behaved as a closed system, and was initially CO_2 -rich, though not CO_2 -saturated, would become increasingly more polymerized with decreasing pressure as a result of the reaction:



It is suggested that increased polymerization of the

melt (prior to the loss of a volatile phase from the olivine melilitite magmas) would result in changes in the olivine-liquid distribution coefficients for the transition elements, leading to more Fe- and Mn-rich olivines in equilibrium with a given liquid composition, and that this in part explains the range in magma-cryst chemistry. However, a consequence of increasing polymerization of the liquid would be that equilibria such as:



would be driven to the right as pressure decreases, and an increasing proportion of CO_2 is dissolved as a carbonate. The result would be a progressive decrease in oxygen activity in the magma subsequent to separation from the source area. While oxygen activities remain above the Fe/FeO buffer, these changes would enhance Fe- and Mn-enrichment in the coexisting olivine, but lead to lower nickel contents due to the marked lowering of $a_{\text{Ni}^{++}}$ in the melt. (Note that oxygen activities are higher for the Ni/NiO buffer than for the Fe/FeO buffer at a given temperature and pressure, Fig. 3.8.) In other words, decreasing oxygen activity would compensate for the increase in the olivine-melt partition coefficient for Ni expected as a result of an increasing degree of polymerization of the magma.

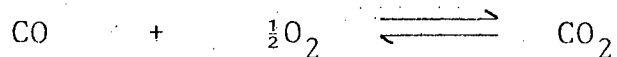
- (f) Summary. The foregoing considerations make it possible to summarize the overall scheme for the evolution of

olivine melilitite magmas and compositions of coexisting olivines as follows:

1. A CO_2 - and carbonate-rich magma is generated in equilibrium with highly magnesian (Fo_{92}) olivines in the source area. Such a model is in accordance with that envisaged by Brey and Green (1977). Initial olivines crystallizing from the magmas should be identical in composition to those in the source area.
2. Progressive polymerization of the magma with decreasing pressure as dissolved CO_2 is converted to carbonate results in an increase in olivine-melt partition coefficients for the transition elements, and at the same time, a marked lowering of oxygen activity. The megacryst suite crystallizes under such changing physico-chemical conditions with olivine compositions changing towards those represented by the HILN compositions. It is implicit that the magma behaved as an essentially closed system during this stage of evolution.
3. With decreasing pressure, the magma would become saturated with respect to CO_2 , and this would initiate the separation of an immiscible carbonate liquid and, particularly at lower pressures, a CO_2 -rich vapour phase. Loss of CO_2 would probably be accompanied by a depolymerization of the liquid as a result of reactions such as:



It is suggested that the resultant structural changes to the liquid would reverse the sense of changing partition coefficients for the transition elements. In addition, the equilibrium reaction:



would be driven to the left, resulting in an increase in oxygen activity. These processes might be partially responsible for the reversed zoning of the HILN olivines, though are difficult to distinguish from those related to late-stage equilibration towards hopper margin compositions. The reversed zoned ilmenites in kimberlites which Haggerty (1975) describes may be related to similar processes.

4. At low pressures (30-40 km to the surface) carbonate solubilities decrease rapidly (Mysen et al., 1975) and volatile loss is likely to be high. Decreasing pressure would therefore result in progressively greater degrees of olivine supersaturation in the liquid and olivines would begin to develop hopper growth forms. Under these conditions there would be rapid changes in partition coefficients and simultaneously a marked increase in oxygen activities. The interaction of these two effects would determine the core composition of the "unusual" hoppers.
5. Increasingly greater relative changes in pressure

at shallower levels would further increase the rate of volatile loss. Olivine crystallization rates would increase and hopper morphology become more pronounced. As a result of large-scale olivine formation, the effects of Raleigh fractionation would be superimposed on those resulting from changing partition coefficients. This would control the variation in composition of the cores of the "normal" hoppers which comprise the dominant olivine population.

6. Late-stage crystallization of titanomagnetite favoured by high oxygen activities results in reversed zoning of the hopper margins. Margins of early-crystallizing olivines equilibrate towards ground-mass composition.

From the foregoing summary, it follows that the olivine populations described form part of a continuous and related evolutionary sequence. The differentiation of the olivines into chemically and petrographically distinct populations may in a sense be somewhat artificial but is, however, useful for descriptive purposes, since the general petrographic and chemical distinctions described earlier were made prior to the interpretations given above.

- (g) Zwartheuwel euhedral olivines. Chemical zonation patterns in euhedral olivines from the (megacryst-bearing) Zwartheuwel pipe differ in detail from those previously

described for several other pipes. However, it will be shown that such zonation patterns can be readily understood in terms of the general model proposed to explain olivine chemistry in the Namaqualand volcanics.

The euhedral olivines analysed from the Zwartheuwel pipe are from a hand specimen (ZH-14) containing the large megacryst illustrated in Plate 17. Olivine chemistry is illustrated in Fig. 3.19. The cores of euhedral olivines differ markedly one from another in terms of both $Mg/(Mg + Fe)$ and Ni contents and there is no rational pattern to these variations. The edges collectively define a linear trend of decreasing Ni and slightly decreasing $Mg/(Mg + Fe)$. The sense of zonation from centre to margin of individual euhedral olivines is always towards the line defined by the crystal edges. This in most olivines results in an initial outward increase in $Mg/(Mg + Fe)$ and Ni followed by iron enrichment and Ni depletion at the very edge of the crystals. A few individuals however show a decrease in $Mg/(Mg + Fe)$ from the centres outwards.

It is impossible to ascribe the marginal increase in Ni concentration in the euhedral olivines to any model involving simple Raleigh-type fractionation. Ni^{++} is preferentially incorporated in olivine relative to equilibrium basaltic liquids and crystallization of the phase would be expected to deplete the residual liquid in Ni. It is therefore proposed that the euhedral olivines in

ZH-14 could all be regarded as being chemically equivalent to "unusual" hoppers and that the zonation patterns were initially determined largely by the interaction of increasing oxygen fugacity and decreasing $K_{\text{Mg-Fe}}^{\text{ol-liq}}$ during magma ascent, rather than the effects of Raleigh fractionation. In other words, the outward increase in Ni concentration in many olivines was probably largely controlled by an increase in oxygen fugacity with progressive crystallization, whilst the reversed zoning with respect to $\text{Mg}/(\text{Mg} + \text{Fe})$ was probably influenced by the combined effects of increasing oxygen fugacity and decreasing $K_{\text{Mg-Fe}}^{\text{ol-liq}}$ during ascent and volatile loss from the magma. The trend of decreasing Ni and $\text{Mg}/(\text{Mg} + \text{Fe})$ which is defined by edge compositions could be interpreted in terms of the effects of Raleigh-type fractionation subsequently overriding those related to changes in oxygen fugacity and $K_{\text{Mg-Fe}}^{\text{ol-liq}}$.

It follows from this interpretation that olivine chemistry and zonation patterns are determined by the interplay of several independent controls. The relative importance of these different factors in determining olivine compositional variation might vary from pipe to pipe as a function of magma composition, ascent velocities and therefore rates of volatile loss. These in turn would influence changes in oxygen fugacity and partition coefficients and determine the importance of Raleigh fractionation effects. Thus, slow rates of ascent and volatile loss might favour slower crystallization and possibly result in changes in oxygen fugacity

and partition coefficients overriding the effects of Raleigh crystallization. On the other hand, rapid ascent of a Mg-rich (olivine saturated) liquid might lead to extremely rapid olivine crystallization rates, thus exaggerating the importance of a Raleigh fractionation control.

The zonation patterns in the euhedral olivines in ZH-14 are of considerable importance in interpreting the origin of the olivine melilitite magmas. Ni concentrations (up to 0.37% Ni) near the margins of some of these olivines are similar to or slightly higher than those found in mantle-type olivines (e.g. Emeleus and Andrews, 1975, Table 2; Nixon and Boyd, 1975, Table 1). This suggests that, at least with respect to Ni content, a liquid composition equivalent to ZH-14 may have been approximately in equilibrium with mantle olivines - or in other words, that such a magma had suffered very little relative Ni loss as a result of crystal fractionation during ascent and crystallization. Unfortunately, as the model for the evolution of the Namaqualand magmas involves changes in partition coefficients for the transition elements during ascent, it is not possible to quantitatively determine the extent of Ni loss - if any. The most fosteritic euhedral olivines in ZH-14 ($\text{Fo}_{89.7}$) fall within the range of iron-rich olivines of presumed mantle derivation (Kesson, 1973). This may indicate that the host magma underwent little fractionation with respect to Mg/Fe subsequent to separation from the source area. Unfortunately, the exact composition of source area olivines is uncertain and, as with the Ni concentration, the Mg/Fe ratio of the olivines was probably controlled by changing partition coefficients during

magma ascent. In addition, crystallization of the (iron-rich) cores of these olivines would lead to a relative depletion of the residual magma in Mg, resulting in a lower fosterite content at the edges than might otherwise be expected. Thus the compositions of ZH-14 euhedral olivines offer intriguing indications that they may have crystallized from an essentially primitive (primary) magma, although the evidence is unfortunately equivocal.

One of the olivines in ZH-14 was identified petrographically as belonging to the HILN population. This is shown by the diamond ornament in Fig. 3.18. The centre of this olivine has the highest Fe and lowest Ni content of those analysed, confirming the petrographic identification. (Note that HILN olivines have been defined as being enriched in iron and depleted in Ni relative to the euhedral olivines in the same rock. The term does not imply any absolute Ni or Fe contents.) It is however significant that the HILN olivine in ZH-14 lies on the trend of decreasing $Mg/(Mg + Fe)$ and Ni defined by the megacrysts from the same pipe, and on chemical grounds could therefore be regarded as being part of the megacryst suite. This emphasises the fact that the olivines from the Namaqualand pipes constitute a chemically continuous and related population, and that the separation of the olivines into different groups, although useful for descriptive purposes, is rather artificial in terms of chemical variations.

3.2.2.5 *Olivine Morphology and Crystallization Rates*

Donaldson (1976) demonstrates that olivine morphology is systematically related to the MgO content and the cooling rate or, alternatively, the degree of supercooling of the parent liquid. Thus, a comparison of the olivine growth forms produced experimentally by Donaldson (op. cit.) with those found in the Namaqualand volcanics should give an indication of either the cooling rates or the degree of supercooling that may be applicable to these magmas.

Olivine morphology and size show no systematic relationship to the relative coarseness of the groundmass assemblage in the Namaqualand volcanics. It is thus very unlikely that the hopper growth forms reflect rapid cooling rates associated with the emplacement of the magma in the upper crust. The preferred explanation for rapid crystallization is a rise in liquidus temperatures associated with a loss of volatiles and an immiscible carbonate liquid during magma ascent, as discussed in Section 3.2.2.4.

Donaldson (op. cit.) points out that comparable crystal shapes grow at the same degree of supercooling irrespective of the melt composition. Consequently, the olivine growth forms produced during isothermal crystallization runs on an Apollo 12 basalt (11.7% MgO) were used for comparison with those found in the Namaqualand volcanics. Olivines from three samples from the southern pipe cluster (Gem-1 (9.9% MgO); Dik-9 (12.6% MgO) and Bies-1 (14.0% MgO)) were compared with growth forms produced experimentally by Donaldson (op. cit.). In

each of these samples, olivines show a range in morphologies from essentially non-skeletal polyhedral individuals through hoppers with pronounced re-entrants and hollow interiors to linked parallel growth forms (see Plates 7, 8 and 10). These correspond to those produced by supercooling of some 10-30°C (or alternatively, cooling rates of 2-40°C/hour for compositions equivalent to the Apollo 12 basalt).

Olivines from three samples from the northern diatreme cluster (WK-1 (17.6% MgO); Ho-1 (21% MgO) and ZHM-3 (24.5% MgO)) were compared with the growth forms produced experimentally by Donaldson.

In WK-1 (Plate 9), there is once again a range in morphology from polyhedral forms to linked parallel growth forms. However, the latter are rare, skeletal habit is far less pronounced in the majority of the olivines in comparison with those in the southern (MgO-poor) volcanics and there are markedly fewer large hopper olivines. The range in morphologies, as before, indicates supercooling of roughly 10-30°C. It is significant that in cooling rate crystallization runs on an olivine eucrite with a magnesian content (18.6% MgO) comparable to that of WK-1, hopper olivines were not produced - only chain and lattice forms, even at the slowest cooling rates (Donaldson, *op. cit.*, Table 3a). This supports the argument that growth forms in the Namaqualand volcanics result from varying degrees of supercooling rather than rapid cooling rates.

Hopper growth forms are poorly developed in the most magnesian

specimens studied (e.g. BKB-1, BKB-2, ZHM-3, ZHM-4, ZHM-5) and most of the olivines in these samples are either polyhedral or slightly skeletal. Furthermore, they are never as large as the largest hoppers found in the southern diatremes. The morphologies indicate either supercooling of less than 10°C or alternatively, cooling rates below 3°C/hour (see Donaldson, Fig. 8 and Table 4).

From the comparisons made above, it is apparent that the hopper olivines have morphologies which indicate a range of supercooling for each rock. This could be explained if the rate of volatile loss and hence the degree of supercooling increased during magma ascent. Mysen et al. (1975) show that CO₂ solubility in basic melts is strongly pressure dependent. Their experimental runs at 1 650°C with a melt of olivine nephelinite composition show a pronounced relative decrease in CO₂ solubility from 4.6 weight % to about 1 weight % in the pressure range 20 Kb to 5 Kb. The relative decrease in solubility from say 30 Kb to 20 Kb (7 weight % to 4.6 weight %) is much smaller.* The relative proportions of volatiles lost and therefore the degree of supercooling would be expected to be greatest at low pressures.

3.2.2.6 *Depth of Hopper Olivine Crystallization*

A crude estimate of the depth of hopper olivine crystallization can be made from textural evidence and theoretical considerations:

It has been suggested (Section 3.2.1) that crystallization of

* Note: With CO₂ contents of less than 4.6% CO₂, appreciable vapour loss would not be expected at pressures above 20 Kb.

melilite in the Namaqualand rocks was probably restricted to depths shallower than 30-45 km. In rocks with a fine-grained groundmass, this mineral shows a flow alignment about skeletal olivine phenocrysts, which therefore clearly crystallized before the melilite. This in turn indicates that crystallization of the hopper olivines was initiated at pressures outside the melilite stability field - in other words, possibly at depths in excess of 30-45 km. This evidence is not definitive unfortunately, for the depth of melilite crystallization in natural systems cannot be accurately fixed, but only very loosely bracketed.

Experimental evidence (Mysen et al., 1975) discussed in the previous section, indicates that there is a large relative decrease in the solubility of CO_2 in basic igneous melts at pressures below 20 Kb. The relative proportions of volatiles lost from the olivine melilitite magmas would therefore be expected to be high at pressures less than 20 Kb. As the growth of hopper olivines has been ascribed to the loss of volatiles from the parent magma, it is plausible to suggest that crystallization of the phase commenced at depths somewhat shallower than 60 km (i.e. pressures less than about 20 Kb). This conclusion is in broad agreement with that made on the basis of textural relationships between olivine and melilite. However, the uncertainties involved in both approaches are such that the inferred depth of crystallization should only be regarded as a very crude estimate.

3.2.2.7 *Alteration to Olivines*

Alteration of olivines, when present, is generally either to green bowlingite or brown iddingsite. In fresh samples, this is usually confined to margins and cracks. When both alteration "minerals" are present, the iddingsite forms a rim about the bowlingite, and appears to represent a more advanced stage of a continuous alteration process. Olivines are occasionally replaced marginally by phlogopite, particularly in coarse-grained rocks. "Phlogopitization" of the olivine probably reflects a scavenging of Mg and Fe from that mineral by residual fluids rich in K and H₂O.

3.2.3 *OXIDES*

The textural relationships between ilmenite, titanomagnetite and perovskite are believed to mirror the same physico-chemical changes to the Namaqualand magmas that are recorded by the complex olivine chemistry. These oxides are therefore conveniently discussed together.

3.2.3.1 *Ilmenite and Titanomagnetite*

Ilmenite is rare in the Namaqualand volcanics, though has been identified in several of the pipes studied. The limited microprobe data available (Table 2(p)) indicates that these ilmenites are relatively enriched in MgO (up to 11.6% MgO) and therefore resemble the picroilmenites recovered from kimberlites (Haggerty, 1973 and 1975), although chromium contents are rather low (0.08% to 0.38% Cr₂O₃). The ilmenites in the Namaqualand volcanics, when present, are invariably

surrounded by complex reaction coronas which all show the same mineral zonation sequence. This illustrated in Plates 24 and 25. The central ilmenite core appears always to grade into a rim of angular titanomagnetite fingers, that are commonly intergrown with a colourless mineral - apparently a zeolite. This rim is always armoured by a narrow perovskite rind, which is in turn invariably surrounded or partially surrounded by an outer clustering of titanomagnetite microphenocrysts. The origin of the ilmenite is equivocal, but it is suggested that the mineral is a cognate phenocryst phase, and crystallized under reduced conditions analogous to those invoked to explain the HILN olivine chemistry, and further, that the rim of skeletal titanomagnetite fingers about ilmenites records the same increase in oxygen activity that led to the crystallization of the hopper olivines. In other words, rapid crystallization, as in the case of the hopper olivines, followed volatile-loss and a rise in the oxygen activity of the magma.

Titanomagnetite ranges in size from phenocrysts, up to 10 mm in diameter, down to a groundmass dusting, indicating that the mineral, like olivine, had an extended period of crystallization. The larger phenocrysts may sometimes be rounded and embayed, suggesting resorption, though angular blocky margins are more common and some show skeletal forms (Plate 27) - probably analogous to the growth features that characterize the hopper olivines. In the southern (Mg-poor, Fe-rich) volcanics, titanomagnetite microphenocrysts are often marginally enclosed by olivine phenocrysts, indicating that the period of crystallization of the two phases partially overlapped.

There is often an unusual association between titanomagnetite and zeolites, which may take on a variety of apparently related forms: the latter minerals are sometimes enclosed by large titanomagnetite phenocrysts, while zeolite-rich segregations in the olivine melilitites such as those described in Section 3.3 may be partially or wholly enclosed by a rim of titanomagnetite microphenocrysts. A particularly unusual association of these phases is illustrated in Plate 26. This has the appearance of a titanomagnetite-zeolite cotectic intergrowth surrounded by a partial rim of later titanomagnetite. The zeolites in the intergrowth have been optically identified as thompsonite (rectangular outline) and natrolite (radiating needles). The significance of this association is discussed in Section 3.3.

Titanomagnetite analyses are presented in Table 2(p). The most notable feature is a marked increase in Ti concentration from centres to edges of the phenocrysts. Groundmass microphenocrysts are also enriched in Ti relative to phenocryst centres.

3.2.3.2 *Perovskite*

Perovskite typically occurs as two distinct populations in the olivine melilitites. The earlier of these shows a rather restricted size range (0.02 - 0.16 mm) and comprises irregular anhedral, rude cubes and interpenetrant twins. Such microphenocrysts are sometimes partially enclosed by olivine in rocks from the southern (though not the northern) pipe clus-

ter. They are therefore believed to represent a primary near-liquidus population. Sugary anhedral in the groundmass represent a distinctly later perovskite population that probably crystallized during emplacement and rapid cooling of the magma. Perovskite has also been found as a narrow rim surrounding a spinel harzburgite inclusion recovered from the pipe code-named Jen. Reid et al. (1975) report similar rims mantling olivines from the Ingwisi Hills diatremes. It is unlikely that these features indicate reaction relationships, and the perovskite appears merely to have crystallized about favourable nuclei.

A very unusual banded perovskite-rich inclusion has been recovered from the Zwartheuwel pipe (Plate 28). The lowermost layer of the inclusion (as illustrated) is dominated by elongate perovskite aggregates that appear to have grown radially outwards from a nucleating surface that is no longer preserved. Titanomagnetite, nepheline, olivine and perovskite are interstitial phases. These latter minerals become more abundant towards the top of the first layer, which is marked by an abrupt change in modal mineral proportions. Elongate perovskite aggregates, growing sub-perpendicular to this boundary surface, dominate the second layer. These aggregates are somewhat shorter than those in the first layer. Titanomagnetite, nepheline, olivine and equant perovskite anhedral are once again interstitial phases. The second layer is overlain by a narrow band rich in titanomagnetite, followed by a third zone dominated by elongate perovskite aggregates (short-

ter than those in the previous two), a second narrow band rich in titanomagnetite and, finally, a layer dominated by intergrown clinopyroxene and nepheline (which is not shown in Plate 28). The clinopyroxene sometimes occurs as branching forms that are believed to be rapid growth features analogous to the branching olivines described by Donaldson (1976). The unusual radiating habit of the perovskite in the inclusion is believed to be analogous to the "comb layering" described by Drever and Johnson (1957), and similarly related to a rapid growth origin.

The minerals present in the inclusion are all common to the olivine melilitites. Those in the lowermost layer are relatively early-crystallizing phases in these volcanics, while those of the final layer are equivalent to the groundmass assemblages. The layering thus mimics the crystallization sequence of the host magma, and there seems little doubt that the inclusion is cognate rather than accidental. It is therefore suggested that the textural features described record the same changes to the magma that initiated rapid growth of the hopper olivines. The nature of the nucleating surface for the inclusion remains speculative, but may have been the walls of the conduit or feeder channels to the magma at depth. The mineralogical layering in the inclusion is probably controlled by the interaction between crystallization and ionic diffusion rates as described by Drever and Johnson (op. cit.). Rapid crystallization of one phase (for example perovskite) relative to ionic diffusion rates might lead to supersaturation of the magma envelope adjacent to the nucleating surface by a second phase (titanomagnetite), which might in turn undergo rapid

crystallization. Saturation with respect to the first mineral might again occur, leading to renewed rapid crystallization of that phase.

Textural relationships shown by the ilmenite-titanomagnetite-perovskite reaction rims described previously indicate that ilmenite was the earliest-crystallizing Ti-rich phase. The reason for the initial suppression of perovskite stability is uncertain. One possibility however, is that complexing between Ca^{++} and CO_3^{--} ions in a carbonate-rich magma resulted in a decrease in Ca^{++} activity, and that this, coupled by the high Fe contents and low oxygen activity of the olivine melilitite magmas, resulted in ilmenite initially being the stable Ti-bearing phase. Loss of an immiscible carbonate phase and volatiles, and the subsequent increase in Ca^{++} and oxygen activities may have resulted in perovskite replacing ilmenite as the stable Ti-rich phase.

3.3 LATE-STAGE IMMISCIBLE LIQUIDS

Gerrard (1958) has described what he terms "ijolite pegmatites", that occur as rare irregular segregations associated with the olivine melilitites of Sutherland and the southern Cape Province. Sick (1970) reports finding a single leucocratic rock ("*das hellen Gestein*") associated with the Sternberg olivine melilitite, Federal Republic of Germany. Sick (op. cit.) terms this leucocratic rock a pyroxene nephelinite, and from his description, it appears closely to resemble the pegmatites described by Gerrard (op. cit.). Similar pegmatites (originally reported by Rogers, 1911) have been found

associated with several of the Namaqualand olivine melilitite pipes. They are usually found only as isolated boulders though in two of the largest pipes (Zwartheuwel and Hoodkop) occur as small irregular outcrops, a few metres across. Occasionally, pegmatite veinlets a few centimetres across, are found cutting olivine melilitite boulders. These veins commonly have a narrow fine-grained chill zone at the contact with the host rock.

The pegmatites have essentially the same mineralogy as the groundmass assemblage of the olivine melilitites (i.e. clinopyroxene, interstitial nepheline and zeolites, with accessory titanomagnetite, apatite and phlogopite). Although coarse-grained, they have petrographic characteristics which indicate rapid crystallization: devitrified glass is present in some, and the apatite commonly occurs as hollow needles with a high length to breadth ratio (up to 54, Plate 30). Wyllie et al. (1962) have demonstrated experimentally that such skeletal apatite morphology is typical of quench textures. The perovskite in the pegmatites sometimes occurs as elongate aggregates along grain boundaries of other phases, particularly clinopyroxene (Plate 30). This will be referred to as "dog-tooth perovskite" because of its serrated appearance and resemblance to a row of dog's teeth. This unusual morphology may also be related to rapid crystallization of the pegmatite. Gerrard (1958) and Sick (1970) have interpreted the pegmatites as being late-stage differentiates of the olivine melilitites. In order to place more rigorous constraints on the

origin of these rocks, two pegmatite samples (ZH-8 and ZH-10), both from the Zwartheuwel pipe, have been analysed. Table 4 gives the mean composition of these two specimens and also a mean composition of the remaining Zwartheuwel samples (excluding ZHM-3, 4 and 5, which are believed to represent a separate intrusion). These data do not lend support to a simple differentiation model for explaining the origin of the pegmatites. The chief difficulty is that the pegmatites are silica-enriched relative to the olivine melilitites. However, olivine, which is the dominant phenocryst phase in the latter rocks, is silica-rich relative to the average Zwartheuwel bulk rock composition (and in fact relative to all the Namaqualand olivine melilitites). Crystallization of this phase would therefore deplete residual liquids in SiO_2 . The same would apply to the crystallization of clinopyroxene, nepheline and melilite. These latter phases are in any event typically present in the groundmass assemblage of the olivine melilitites and are therefore unlikely to have controlled the evolution of liquid compositions.

Silica enrichment could in theory have been effected by large-scale fractionation of titanomagnetite and perovskite, which appear to have been near-liquidus phases. However, removal of titanomagnetite would be expected to rapidly deplete residual liquids in Fe and trace elements such as V. Fe is not markedly depleted in the pegmatites and these rocks are in fact relatively enriched in V. Similarly, fractionation of perovskite would deplete residual liquids in TiO_2 , CaO and

probably Zr (data presented by Smith, 1970, indicate that the partition coefficient for Zr between perovskite and ultrabasic magmas will be slightly greater than unity). Relative to the olivine melilitites, the pegmatites are enriched in Ca and Zr and only marginally depleted in Ti.

Such considerations strongly suggest that the pegmatites cannot be related to the olivine melilitites by crystal fractionation processes. A further drawback to such a model is that compositions intermediate between the two rock types appear to be absent. It is therefore proposed that the pegmatites and olivine melilitites represent complementary immiscible liquids which evolved from a homogeneous parent magma. Several lines of chemical evidence can be led to support such a model:

Column 3 of Table 4 gives concentration ratios for different elements in the pegmatites and average Zwartheuvel olivine melilitite. These ratios, which could be regarded as crude partition coefficients for the two hypothetical liquids, indicate a fairly coherent distribution pattern. MgO, and the transition elements Fe, Mn, Ti, Zn, Ni, Co and Cr are relatively enriched in the olivine melilitites. Common to all of these elements is a tendency to form minerals with relatively unpolymerized mineral structures. On the other hand, K, P, Si, Al, Sr, Rb and Ba, which tend to favour highly polymerized mineral structures, are relatively enriched in the pegmatites. Such a geochemical distribution pattern is consistent with that expected for coexisting liquids with different

structures.

The distribution pattern is also qualitatively similar to that found in coexisting glass inclusions trapped in late-crystallizing minerals in lunar rocks (Roedder and Weiblen, 1970 and 1971). These authors distinguish between high- and low-silica glasses which they interpret as complementary immiscible liquids. The former are enriched in Si, Al, Na and K, and probably Sr, Ba, Rb and Zr. With the exception of Na, these elements are enriched in the pegmatites relative to the olivine melilitites. Fe, Mg, Ca, Ti and probably Mn and P are relatively depleted in the high silica glasses. With the exception of Ca and P, all of the latter are relatively depleted in the pegmatites. Such qualitatively similar distribution patterns support the possibility that the pegmatites and olivine melilitites, like the lunar glasses, represent coexisting immiscible liquids. The significance of the aberrant behaviour of Ca, P and Na is uncertain, though it is noted that Na concentrations may have been influenced by liquid-vapour partitioning (Section 4.4.5), while the distribution of the former two elements may have been controlled by equilibria with an immiscible carbonate phase (Section 4.4.4).

Gélinas et al. (1976) point out that the compositions of glass inclusions in lunar rocks tend to plot at opposite ends of the field of immiscibility outlined by Holgate (1954) in the pseudo-ternary system $\text{SiO}_2 - (\text{CaO} + \text{MgO} + \text{FeO} + \text{TiO}_2) - (\text{Na}_2\text{O} + \text{K}_2\text{O} + \text{Al}_2\text{O}_3)$. The olivine melilitites plot close to the field of lunar rocks in this system, and are clearly se-

parated from the pegmatites (Fig.3.20), which are displaced towards silica-rich compositions close to but outside the field of immiscibility defined by Holgate (op. cit.). This emphasises the marked compositional gap between olivine melilitites and pegmatites, and lends support to the likelihood that the two rock types represent complementary immiscible liquids.

In thin sections of the Namaqualand volcanics, it is possible to recognize a variety of different mineral segregations that are believed to be genetically related to the ijolite pegmatites. Very coarse-grained olivine nephelinites sometimes enclose rounded "pools", ranging from less than 1 mm to a few centimetres in diameter, that are composed of essentially the same minerals as the surrounding groundmass, but are markedly coarser-grained (Plate 31). The edges of such "pools" are often marked by a narrow chill zone composed chiefly of clinopyroxene needles intermediate in length between those of the surrounding groundmass, and the central coarser part of the "pool". The morphology of these segregations is consistent with their having crystallized from an immiscible liquid. It is suggested that they are small-scale equivalents of the ijolite pegmatites, which they closely resemble mineralogically.

Apparently related petrographical features that are common to all of the olivine melilitites, are small irregular stringers and veinlets or patchy segregations in the groundmass (Plate 29). These are always dominated by varying proportions of zeolites and clinopyroxene, often showing a zonal mineral

distribution. This is commonly from a fine-grained outer margin composed chiefly of tiny clinopyroxene needles, to an inner zone of coarser clinopyroxene laths, which in turn surrounds a core composed principally of a mixture of zeolites - chiefly natrolite and thompsonite. Titanomagnetite, perovskite, phlogopite, apatite and pyrite may be present in varying proportions as accessory phases, particularly in the larger and coarser segregations. Calcite is occasionally present as irregular patches, or replacing zeolite (natrolite?) needles, but is atypical.

There is a continuous range in modal mineral proportions, and thus also in composition, from irregular zoned zeolite-rich segregations and rounded "pools" to coarse pegmatites that are dominated mineralogically by clinopyroxene and zeolites, but also include a variety of different accessory phases (perovskite, titanomagnetite, apatite and phlogopite). It is suggested that such a range in modal mineral proportions records the continuous evolution of an immiscible liquid from the host olivine melilitite during cooling. The most zeolite-rich segregations probably reflect the presence of a hydrous and alkali-rich fluid phase - possibly a vapour - that evolved at relatively high temperatures, was trapped and subsequently crystallized at low temperatures. With sufficiently slow cooling rates, it is suggested that such a fluid would change in chemistry towards compositions represented by the ijolite pegmatites. Judging from their mineralogy (i.e. the greater proportions of clinopyroxene, titanomagnetite and

apatite), the latter would be enriched in Mg, Fe, Ti and Ca relative to the early-formed fluids.

The quench textures of the ijolite pegmatites and the fine-grained clinopyroxene rims associated with many of the segregations indicates that crystallization of the immiscible liquids was often rather rapid. This may be related to rapid loss of volatiles from these liquids during emplacement.

In the Hoedkop pipe, phlogopite is concentrated as irregular blebs and stringers close to and roughly concentric with the pipe margins. These are thought to result from a mechanical segregation of late-stage hydrous fluids during convectional overturn of the magma at the pipe margins (see Section 2.1.2). Such phlogopite-rich segregations may also be related to the late-stage immiscible liquids.

Late-stage zeolite-rich veins fill tension joints in the Dikdoorn pipe. They apparently represent a fluid phase associated with the waning phases of igneous activity in this pipe.

Titanomagnetite-zeolite intergrowths. An unusual association between titanomagnetite and zeolites has been described in Section 3.2.3.2. It is most likely that the zeolites associated with this opaque phase are related to the evolution of a hydrous pegmatitic fluid. The textural relationships described would therefore seem to be most readily interpreted as indicating that zeolite-rich segregations formed favourable nuclei for titanomagnetite crystallization. This may have been the result of locally higher oxygen activities in the

vicinity of such segregations.

Evidence for four coexisting immiscible fluids. The hopper olivines enclose a variety of different types of inclusions (described in Section 3.2.2.1). On the basis of their morphology, three of these - the zeolite-clinopyroxene bearing composite inclusions and the sulphide and carbonate inclusions - have been interpreted as representing liquids trapped at the time of hopper olivine crystallization. (Note that although the morphology of these inclusions cannot be regarded as conclusive evidence that they were trapped as liquids, this would appear to be the simplest interpretation.) Fluid inclusions with bubbles have been interpreted as having been trapped as a vapour phase.

The abundance of zeolites in the composite inclusions indicates that the trapped liquid was relatively hydrous. This in turn suggests that such a liquid was related to the immiscible pegmatite fluids described in this section rather than the complementary drier olivine melilitite magma. The similar mineral assemblages of the composite inclusions and pegmatites supports this interpretation. It would therefore appear that the following liquids would have been present during all or part of the period of hopper olivine crystallization:

- (a) The host olivine melilitite magma.
- (b) Water-rich pegmatite fluids (ijolite pegmatites) in various stages of development.

(c) A carbonate liquid.

(d) A sulphide liquid.

The different types of inclusions described have not all been found coexisting in a single hopper olivine. It is therefore uncertain whether the various hypothetical liquids coexisted at any stage during the evolution of the olivine melilitite magmas. Although such a situation might on superficial consideration appear implausible, the possibility should not be ruled out however, and it is interesting to consider the factors which may have led to the development of four coexisting liquids.

In Section 3.2.2:4, it was suggested that CO_2 and an immiscible carbonate liquid may have been lost from the host olivine melilitite magma during ascent, and that this would have led to marked structural changes to the parental magma. If so, such changes may in turn have triggered the separation of an immiscible pegmatite liquid and a sulphide liquid to accommodate those components which became insoluble in the decarbonated (and consequently relatively depolymerized) olivine melilitite magma.

3.4 PETROGRAPHY OF ULTRAMAFIC XENOLITHS

Ultramafic xenoliths are not common in most of the Namaqualand olivine melilitites and olivine nephelinites, though are present in appreciable amounts in the Hoedkop and Zwartheuvel pipes. The xenolith suite has not yet been extensively stu-

died, but a cursory investigation indicates that harzburgites are always the most abundant of these accidental inclusions. Rare garnet lherzolites have been found in the Hoedkop pipe, but not any of the others studied. A single five-phase garnet-spinel lherzolite with very pale mauve garnets was also found in the Hoedkop pipe. The ultramafic xenoliths are all relatively small, and seldom exceed 10 cm in greatest diameter.

The garnet lherzolites would be termed common peridotites according to the classification scheme of Gurney et al. (1975), and coarse equant peridotites according to the scheme proposed by Hart (1977) (see Plates 32 and 33). Olivine is always the most abundant phase present, and characteristically has smoothly curving and occasionally straight edges. Some of the olivines show slight undulose extinction under crossed nicols. They generally range in size from 1.2 to 8 mm across, although occasionally, recrystallization has produced zones of small (0.1 mm) equant olivines. Garnets occur sporadically as rounded grains, up to 4 mm in diameter, that are invariably surrounded by a kelyphytic rim with abundant brown spinel anhedral (0.08 mm in diameter). Clinopyroxene and orthopyroxene generally have smoothly curved margins, and are typically smaller than the other two phases (1-1.5 mm). One small rounded clinopyroxene grain (0.08 mm in diameter) was found wholly enclosed by garnet in one of the garnet lherzolites. Phlogopite occurs along cracks and about clinopyroxene margins in some, though not all of these xenoliths. When present, the

phase is considered to be secondary.

Mineral analyses for one of the garnet lherzolites (Ho x) are given in Table 2(o). The minerals have compositions that are broadly similar to those in garnet lherzolites from kimberlites. However, in comparison to the latter, garnets in xenoliths from the olivine melilitites are somewhat richer in Ca and poorer in Mg relative to Cr contents (Gurney and Switzer, 1973) while the clinopyroxene gives a pressure of 38 Kb based on the uncorrected Al content (MacGregor, 1974). Using the procedure outlined by Fraser and Lawless (1978), equilibrium at 937°C and 25 Kb is indicated. This suggests derivation from shallower depths than the source of kimberlitic garnet lherzolites.

The harzburgites are texturally similar to the garnet lherzolites. Some of these rocks have accessory red-brown spinel anhedral (0.25 mm in diameter).

Because of the small size of the ultramafic xenoliths, reliable estimates of modal mineral proportions are not possible.

4. BULK ROCK CHEMISTRY

4.1 GENETIC RELATIONSHIP BETWEEN THE NORTHERN AND SOUTHERN PIPE CLUSTERS

Several different varieties of volcanic pipes can be distinguished in the Gamoep-Garies areas on the basis of field characteristics. Temporal relationships are not well established, and it is therefore uncertain whether volcanism was episodic or continuous in Namaqualand during the early/middle Tertiary Period (Section 1). Such uncertainty complicates the interpretation of the bulk rock chemical data, for there may not necessarily be a genetic relationship between rocks of radically different ages. Nevertheless the extremely alkaline, undersaturated nature of the rocks in both pipe clusters and their gross mineralogical similarities suggest a common kinship. Moreover, there appears to be a uniformity in the geological processes controlling the chemistry of magmas erupted in similar tectonic environments. This is illustrated by the remarkable chemical uniformity of many MOR basalts of different ages (Engel *et al.*, 1965) or of many typical continental tholeiites (Walker and Poldevaart, 1949). The two Namaqualand-Bushmanland pipe clusters are found in broadly similar tectonic environments (close to the continental margin in cratonic areas that have been relatively stable during the past 1 b.y.). If tectonic setting is a fundamental factor governing magmatic processes, it is possible that the Namaqualand-Bushmanland volcanics are related in terms of a common geological control or controls. They will therefore be treated as a single chemically related suite of rocks, and one of the objects of this work is to demonstrate that such an assumption is valid. It will

also be shown that the olivine melilitites are closely chemically allied to kimberlites, and that compositional differences between the two rock types are largely related to subtle differences in their respective tectonic settings.

4.2 ANALYTICAL TECHNIQUES

Bulk rock analyses were carried out by x-ray fluorescence spectroscopy (XRF) during routine analytical runs in the Department of Geochemistry, University of Cape Town. Major elements (excluding Na) were analysed using the lithium tetraborate fusion method of Norrish and Hutton (1969). Na and trace elements were analysed on pressed powder briquettes. The quality of the data expected is indicated in Tables 13A and 13B. A more detailed discussion of the analytical procedures and standards used in the Geochemistry Department is given by Willis *et al.* (1971) and Willis *et al.* (1972).

4.3 BULK ROCK CHEMISTRY

4.3.1 GENERAL

Major and trace element analyses have been carried out on a total of sixty-nine whole-rock samples from Namaqualand. Sixty of these are either olivine melilitites or olivine nephelinites, twenty-four of which are from the southern (Garies) group of diatremes, and thirty-six from the northern (Gamoep) cluster. These are all extremely fresh. The analyses are presented in Tables 1(a) and 1(b). The remaining nine analyses comprise two ijolite pegmatite samples (ZH-8 and ZH-10), both from the Zwartheuvel pipe in the northern cluster, and seven from pipes classified as "kimberlites" by exploration geologists working in Nama-

qualand-Bushmanland. Some of the latter are considered to be pseudokimberlites (RIEM and KKV samples), while the Bit sample is from a sediment filled pipe. Because of poor field exposures, it is uncertain whether the remaining samples are true kimberlites or not. For simplicity, all are grouped together as "kimberlites". Bit-1 is from the southern pipe cluster, while the remaining samples are from the northern group of pipes. The pegmatite analyses are given in Table 1(d) and those for the "kimberlites" in Table 1(c). In addition to these samples, melilite-bearing rocks from elsewhere in southern Africa were analysed for comparative purposes. They include specimens from the following localities: Klaasvoogds, Robertson District (Table 1(e)); Spiegel River, Riversdale (Table 1(b)); Sutherland (Table 1(g)) and the Klinghardt Mountains, Namibia (Table 1(h)). The latter specimens were kindly donated by Dr J S Marsh.

Brief petrographic descriptions of individual rocks analysed (excluding the kimberlites, which are highly altered) are given in Table 6.

Average concentrations, and concentration ranges of analysed elements in the Namaqualand volcanics (excluding "kimberlites" and "ijolite pegmatites") are listed in Table 3. The composition of an "average" mid-ocean ridge (MOR) basalt is included in the Table for comparative purposes, as this magma type is apparently wide-spread in time and space, is chemically rather uniform and has been considered as a possible primary magma by some authors (Engel *et al.*, 1965). Relative enrichment or depletion of the different elements in the average Namaqualand composition with respect to the average MOR basalt are shown in column 9 of the

table. This indicates that, compared to MOR basalts, the Namaqualand volcanics are strongly enriched in incompatible elements,* MgO, total iron (expressed as Fe_2O_3) and CaO, but are markedly depleted in SiO_2 .

The widest spread in major element concentration is shown by MgO. It is therefore convenient to use this oxide as a basis for comparing the variations of other elements on binary diagrams. Salient geochemical relationships are illustrated in Figures 4.1 to 4.40. Where (a) and (b) plots are given, the former show only data for the Namaqualand volcanics (including the Namaqualand "kimberlites"), and no distinction is made between samples from different pipes. Correlation coefficients (R) and least squares regression lines for each cluster and for all the data collectively (excluding "kimberlites" in each case) are given in these plots. The (b) plots differentiate between samples from different pipes, and in addition illustrate data for a suite of kimberlites from the Kimberley area, South Africa. These kimberlite data are unpublished, and were kindly made available by Drs J J Gurney and C R Clement.

The Namaqualand bulk rock data are discussed in detail where relevant to developing a geochemical model for the origin of the Namaqualand volcanics. Several generalizations are possible however:

* *Footnote: In this work, the term "incompatible elements" will be used for those elements that are strongly partitioned into the liquid during melting of a four-phase garnet lherzolite (e.g. K, Ti, Rb, Ba, Zr, Nb, P). The "compatible elements" will be considered as those which are not strongly enriched in such early liquids.*

- (1) Rocks from the same pipe generally have either a very small compositional spread, with points plotting close together on the binary diagrams, or else, where a large number of samples have been analysed, they define two or more distinct compositional clusters e.g. Figures 4.1(b); 4.2(b); 4.10(b); 4.18(b); 4.19(b). Thus, Zwartheuwel samples (vertical crosses) define three distinct compositional groups. One of the latter consists of specimens (code-named ZHM) from a low, poorly defined knoll forming the northern part of the pipe. This probably represents a separate intrusion. Exposures are unfortunately too poor to establish whether the other two compositional clusters are also from separate intrusions, though this may prove to be the case. The same probably applies to the two BIES populations, which are very clearly separated compositionally.
- (2) There is only a limited degree of overlap in the compositional fields defined by the two pipe clusters. Moreover, rocks (e.g. WKA-1, ZH-1) from the northern pipe cluster which overlap the compositional field of the southern pipes with respect to certain elements - such as Mg and Al (Fig. 4.1(b)) - are always clearly separated from the latter field with respect to other elements - for example, Zr, Nb, Cr, Ca and Zn (Table 3). In general, the northern volcanics are enriched in Mg, Si, Ni, Cr and Cu relative to those in the south, but are relatively depleted in Al, Ca, Fe and several "incompatible" elements (P, Zr, Nb, Sr and Y). There is however, considerable overlap in the concentration ranges of the alkali elements (Na, K and Rb) and also of Ba

and Ti in the two pipe clusters (Table 3). The latter elements, with the exception of Ti, are characterized by a wide and erratic compositional variation across the whole range of MgO concentrations.

- (3) The geochemical behaviour of Zn is rather unusual as this element shows a quantumized distribution in the Namaqualand volcanics. The Garies samples show a limited and non-systematic concentration spread (125 ppm to 157 ppm Zn) about an average value of 146 ppm, while those in the Gamoep cluster define a narrow spread in concentration from 96 to 120 ppm Zn, with a mean concentration of 109 ppm. (Sample WK-7 falls outside this range and within that of the Garies pipes. This sample is anomalously enriched in Cu but in other respects closely similar to the other four WK specimens. The high Zn and Cu concentrations in WK-7 are therefore believed to be due to contamination during sample preparation, apparently caused by a worn brass screw on the mechanical agate mortar.)

Gurney and Ahrens (1973) have previously drawn attention to the limited ranges of Zn concentration in South African kimberlites (Ave = 55 ppm Zn), ultramafic nodules recovered from kimberlites (Ave = 35 ppm Zn), South African olivine melilitites (Ave = 100-110 ppm Zn) and oceanic basalts (Ave = 100-110 ppm Zn). (Erlank and Reid (1974) have subsequently suggested an average of 122 ppm Zn for oceanic basalts.)

The Gamoep rocks therefore have zinc contents that fall within the olivine melilitite field reported by Gurney and Ahrens (*op. cit.*) while the Garies samples define a separate field that is relatively enriched in Zn. The "quantu-

mized" distribution of Zn in the Namaqualand volcanics is extremely unusual, and any model for their origin should offer a satisfactory explanation for this anomalous geochemical behaviour.

- (4) The elements Zr and Al show a fairly consistent decrease in concentration towards more Mg-rich compositions within both pipe clusters, thus mimicing the overall geochemical trends. In general however, geochemical variation patterns within each pipe cluster are poorly defined.

4.3.2 MIXING MODELS

An attempt has been made to model the geochemical variation within each pipe cluster using least squares mixing calculations (mixes) involving different bulk rock compositions and actual or hypothetical phenocryst phases. In addition, mean compositions for the two pipe clusters (Table 3) have been used to model geochemical variation between the two Namaqualand-Bushmanland compositional fields. Mixing calculations were carried out using a computer programme written by Dr A R Duncan of the Department of Geochemistry, University of Cape Town, and based on the procedure outlined by Bryan *et al.* (1969). Mineral phases used in the mixes can be divided into two groups:

- (a) Low pressure phenocrysts and microphenocrysts (hopper olivine, melilite, perovskite and titanomagnetite) (Section 3).

Melilite shows limited chemical variation in the Namaqualand-Bushmanland volcanics, and a mean composition was assumed for this phase. However, hopper olivines and titanomagnetite show wide and complex chemical variations, and the choice of composi-

tions for these phases is therefore rather arbitrary. The olivine composition used ($\text{Fo}_{85.4}$) represents the Mg-rich end of the hopper olivine compositional range. The spinel composition is roughly intermediate for the spread of available analyses. The perovskite composition was arbitrarily chosen from analyses given by Mitchell (1972). As titanomagnetite and perovskite are invariably minor phases in all mixes, the choice of compositions for these minerals is not too critical.

(b) Real or hypothetical high pressure phases. Petrographic and mineral chemistry data have been interpreted to indicate that olivine megacrysts and ilmenite may be high pressure cognate phenocrysts. Clinopyroxene has also been included as a possible high pressure phase, for, despite the lack of evidence for clinopyroxene phenocrysts, mixes for the northern pipes are improved if this phase is included in the calculations.

Because of the range in chemistry of megacryst olivine and ilmenite in the olivine melilitites, choice of compositions for these phases is also rather arbitrary. The olivine used (Fo_{91}) is from the Mg-rich end of the megacryst range, while the ilmenite composition is similar to those of ilmenites analysed in the olivine melilitites. The clinopyroxene composition was arbitrarily selected from the analyses of a clinopyroxene in a four phase garnet lherzolite recovered from the Hoedkop pipe (Table 2(o)).

4.3.2.1 *Southern Pipes*

Table 10(a) gives various mixes for the southern group of pipes. Mix 1 in this table includes the two extreme rock compositions from this cluster (AMJ478 and AMJ281), together with the low

pressure, near liquidus mineral phases (hopper olivine, melilite, titaniferous magnetite and perovskite) that characterize the olivine melilitites. There is a good correlation between actual and predicted concentrations of Fe, Ti, Ca, Si and Mg in the mix. The correlation for Al is not quite as good however, and there is a very poor correspondence for Mn, K, Na and P.

Included in Mix 1 are calculated values for Zr, Nb, Sr, Ba, Rb and Y in AMJ281, obtained by assuming that these elements are excluded by the low pressure phases during crystallization (*i.e.* that the bulk mineral - liquid partition coefficient is effectively zero). The predicted Zr concentration is slightly lower than that observed, but the agreement is nevertheless fairly close. The predicted Nb concentration is slightly higher than observed, but concentrations of Sr, Ba, Rb and Y are grossly higher than observed.

Mix 2 in Table 10(a) is for two samples from the BIES pipe, in which two distinct compositional groupings can be recognized. As in the previous mix, agreement between calculated and predicted values is good for Fe, Ti, Ca, Si and Mg, moderately good for Al, and poor for Mn, K, Na and P. The "incompatible" trace elements show similar relative behaviour to that described for Mix 1.

The good correspondence between actual and predicted concentrations for many elements in these mixes suggests that crystal fractionation involving the low pressure mineral phases may have been largely responsible for controlling chemical variation in the Garies pipes. Significantly, with the exception of titanomagnetite, the relative proportions of the mineral phases required

by the mixes are comparable to modal proportions of these phases in the southern pipes (Table 5). The relatively high titanomagnetite modal proportions relative to those required by the mixes may reflect either an extended period of crystallization of the opaque, late-stage oxidation of the magma, or both.

It is possible to account for some, though not all of the discrepancies between predicted and actual concentrations in the southern mixes:

In Section 3, attention was drawn to the fact that olivine and titanomagnetite phenocrysts frequently enclose zeolites, which were interpreted as having crystallized from a trapped fluid phase. Thus, removal of olivine and titanomagnetite would probably be accompanied by removal of minor amounts of Al-rich zeolites. This would in turn lead to lower Al_2O_3 concentrations than predicted by the mixes - in other words, mixes could be improved if zeolites were incorporated in the calculations. Removal of minor amounts of zeolites in addition to other low pressure phases would also result in a slight improvement between predicted and calculated Zr concentrations.

The poor agreement for Mn could be explained by assuming that this element behaved as a trace component during crystallization, *i.e.* obeyed Henry's Law. Titanomagnetite incorporates relatively large amounts of Mn, while the remaining low pressure phases used in the mixes all contain small but significant Mn concentrations. It is therefore plausible that the overall mineral-melt partition coefficient for these phases would be close to 1. This would in turn account for the closely similar Mn concentrations

in AMJ281 and AMJ478 (0,243 and 0,240% MnO respectively).

The low Nb and Sr concentrations in AMJ281 compared to predicted values (assuming incompatible element behaviour for these elements) could be explained in terms of these elements being strongly partitioned into one or more of the low pressure phases during crystallization. Data presented by Mitchell (1972) shows that kimberlitic perovskite is characteristically Nb-rich (up to 1,5% Nb₂O₅). Fractional crystallization involving this mineral would therefore strongly influence Nb distribution in derivative liquids. Melilite is a possible Sr host, and is likely to have controlled the behaviour of this element in the Garies volcanics.

Appropriate partition coefficients for Nb and Sr could be calculated by assuming that AMJ478 and AMJ281 bear a parent liquid-daughter liquid relationship, linked essentially by Raleigh-type fractionation of low pressure phases. If Sr is assumed to be effectively excluded by all mineral phases except melilite, and Nb by all except perovskite, then, using mineral proportions from Mix 1 in Table 10(a) it can be shown that values of 3,9 for $D_{Sr}^{melilite-liquid}$ and 19 for $D_{Nb}^{perovskite-liquid}$ would account for the concentrations of the two elements in AMJ281. Analogous calculations involving BIES1 and BIES2 would require that $D_{Sr}^{melilite-liquid} = 3.1$ and $D_{Nb}^{perovskite-liquid} = 22,9$. Although these figures are in good agreement, they should only be regarded as upper limit estimations of the partition coefficients. Lower values would be required if non-Raleigh type addition and removal of phases took place - for example a mechanical mixing of liquids and crystals, possibly involving a convective magma overturn during emplacement (Section 2). Nevertheless, the calculated par-

partition coefficients are not unreasonable. Arth (1976) reports values up to 2,9 for the Sr partition coefficient between plagioclase and basic liquids, while the exceptionally high Nb concentrations reported for perovskite (Mitchell, 1972) are consistent with the predicted high partition coefficient for this mineral.

4.3.2.1.1 *Anomalous Behaviour of P, Y, K, Na, Rb and Ba*

Factors controlling the distribution of P, Y, K, Na, Rb and Ba remain somewhat speculative. The effects of alteration are likely to have been negligible, as the rocks are all extremely fresh. It is therefore necessary to explore alternative avenues to account for the behaviour of these elements.

Variations in P and Y may reflect primary source area inhomogeneities, or possibly the effects of apatite fractionation. The latter mineral is normally only present as a groundmass phase in the olivine melilitites, though is occasionally found, together with zeolites, enclosed by the rare titanomagnetite phenocrysts. Although apatite has not been found in the zeolite-bearing inclusions in hopper olivines, it is possible that it has been overlooked. Separation of even minor amounts of apatite-bearing titanomagnetite or olivine would strongly affect the distribution of phosphorous in the Namaqualand-Bushmanland volcanics.

Variations in the remaining elements may be partly related to source area inhomogeneity. However, it has been suggested (Section 3) that the olivine melilitite magmas lost a CO₂-rich vapour and an immiscible carbonate liquid during ascent to the surface. The vapour would be expected to contain appreciable H₂O, and it is possible that elements such as the alkali metals and Ba would

have been preferentially partitioned into such a phase. Appreciable vapour loss might then have been responsible for the apparently erratic behaviour of these elements in the olivine melilitites, as well as the extremely low Na concentrations in some of the Garies samples.

The geochemical significance of liquid-vapour partitioning is unfortunately difficult to evaluate quantitatively, as there are few published data for liquid-vapour partition coefficients, and it is impossible to estimate relative proportions of vapour lost by the magma. Nevertheless, alkali element metasomatism characterizes many carbonatite complexes and, if the olivine melilitites did indeed lose a vapour-phase during ascent, it is plausible to expect simultaneous loss of alkali elements. Experimental evidence for liquid-vapour partitioning is provided by van Groos and Wyllie (1973) and van Groos (1975) who show that in systems involving equilibria between silicate and carbonate liquids and a vapour, Na is strongly partitioned into the latter phase. Huntington (1975, 1976, 1977) reports that alkali metals and halides in Mt Etna sublimates show enrichment factors up to 3 relative to associated lavas. Although these enrichment factors are unlikely to be reliable indicators of liquid-vapour partition coefficients, they do provide further evidence for vapour transport of alkali elements in volcanic processes.

Loss of a carbonate liquid from the olivine melilitite magmas during ascent may have influenced the distribution of elements such as Ba, P and Y. A small carbonate dyke is associated with the Zwartheuwel pipe in the Garies cluster (Section 2). Semi-quantitative analysis indicates that this dyke is Ba-rich (over 2% Ba).

Moreover, barytes is abundant in the vicinity of the Salpeterkop carbonatite, which is closely associated with the Sutherland olivine melilitites. This evidence suggests that the distribution of Ba would be influenced by the separation of a carbonate fluid. The same may have applied to P and Y. Large apatite concentrations are found associated with the Phalaborwa carbonatite complex in the Northern Transvaal, and although their origin is not well understood, they may reflect phosphorous mobility during emplacement of the parent magma. The latter possibility is supported by Fesq *et al.* (1975). Their data show that kimberlite samples from the Bellsbank Fissures, which are unusually carbonate-rich (up to 20,5% CO₂), are relatively Mg-poor (as low as 10,6% MgO) but markedly enriched in Sr (up to 1500 ppm Sr), P (up to 1.54% P₂O₅) and rare earth elements. These authors recognize the carbonatitic affinities of the carbonate phase and state that "*R.E.E. enrichment and fractionation in kimberlite magmas is considered to be dominantly controlled by selective formation and removal of mobile R.E.E.-carbonate complexes in carbonatitic fluids which separate from the silica-richer complementary 'kimberlite' magma during emplacement*". As the rare earth elements show a strong affinity for phosphate minerals (Deer, Howie and Zussman, 1966) it seems reasonable that the behaviour of P may have similarly been influenced by the segregation of a carbonate-rich fluid from the olivine melilitite magmas.

In summary, it would appear that bulk rock geochemical variation in the Garies volcanics can to a first approximation be explained in terms of addition or subtraction of the low pressure phenocryst and microphenocryst phases. This may have ^{entailed} ~~involved~~ either

simple Raleigh-type crystal fractionation, or possibly a mechanical mixing of liquids and crystals during magma ascent and convective overturn. Source area inhomogeneity and loss of vapour and fluid carbonate phases may have strongly influenced the behaviour of the alkali elements and Ba, P and Y.

Within this framework it is possible to explain the behaviour of various trace elements in the Garies volcanics:

The limited range in Zn concentration may indicate that, as for Mn, the average partition coefficient between liquid and near liquidus phases for this element is close to 1. K G Cox (oral communication) notes that a zincian analogue of melilite has been synthesised, suggesting a relatively high melilite-liquid partition_A coefficient for this element. Olivine is likely to discriminate against Zn, and thus removal of this phase together with melilite in some convenient ratio might account for the behaviour of this element in the Garies volcanics.

Crystal fractionation processes dominated by olivine removal will strongly affect Ni concentrations in derivative and cumulus-enriched liquids. An attempt has therefore been made to demonstrate that Ni variation in the southern pipe cluster is consistent with the proposed fractionation model. The following assumptions have been made:

- 1) A liquid with composition equivalent to the average for the Garies bulk rock compositions (Table 3) has been used as a parental magma for the southern pipes. Such a liquid may of course itself be chemically evolved.

2) Low pressure phenocryst phases (olivine, melilite, titanomagnetite and perovskite are subtracted from the parent magma in the proportions given in Mix 1, Table 10(a). For simplicity, it is assumed that mineral phase compositions do not vary. The scatter in the data do not warrant a more sophisticated treatment.

3) Raleigh-type fractionation occurs.

4) The Ni partition coefficient is related to liquid compositions by the expression $D_{Ni}^{olivine-liquid} = \frac{124.1}{MgO} - 2.85$ (H S Smith, oral communication; see also Section 4.4.2). Partition coefficients for the remaining phases are assumed to be constant and are arbitrarily taken to be as follows:

$$D_{Ni}^{melilite-liquid} = 2$$

$$D_{Ni}^{TiMt-liquid} = 0$$

$$D_{Ni}^{perovskite-liquid} = 0$$

Based on the MgO content of the hypothetical parental liquid for the southern pipes, an olivine-liquid partition coefficient and thus a bulk mineral-liquid partition coefficient was calculated. 5% of the bulk mineral extract was then subtracted from the parental liquid and a new (derivative) liquid composition calculated. A new value for the Ni partition coefficient was then determined, and the process repeated. The result is a hypothetical line of liquid descent (Fig. 4.10(d)) that plots close to the Mg-poor bulk rock compositions for the Garies pipes.

Using the appropriate bulk mineral-liquid partition coefficient, the average Ni concentration of the phases in equilibrium with

the hypothetical parent liquid (i.e. average Garies bulk rock composition) were determined. Thirty percent of the bulk mineral extract was then added to this liquid. The resultant variation vector is shown in Fig. 4.10(d). This line is straddled by the relatively Mg-rich bulk rock compositions.

It would appear therefore that while the scatter in the Ni-Mg data do not uniquely constrain models for the chemical evolution of the Garies pipes, these data do appear to be consistent with the proposed low pressure fractionation model.

4.3.2.2 *Northern (Gamoep) Pipes*

Mixing calculations have also been carried out for different bulk rock samples from the Gamoep cluster. Using the two extreme bulk rock compositions (ZHM3 and ZH1 - both from the Zwartheuvel pipe) together with the low pressure phases olivine, titanomagnetite and perovskite, it is possible to obtain a mix with good agreement for the elements Fe, Ti, Ca, Si and Mg but poor agreement for Al (Mix 1, Table 10(b)). It should be noted that the elements Mn, P, K and Na have been excluded from the mixing calculations. However, if included, these elements show very poor agreement between predicted and actual concentrations, analogous to their behaviour in the southern pipe cluster. It should be noted that the relative proportions of the minerals required by the mix are broadly similar to the relative model proportions for the Gamoep pipes (Table 5).

If Zr, Sr and Nb are treated as being incompatible, calculated concentrations for the former two elements in ZH1 are somewhat lower than those observed, while the predicted Nb concentration is

slightly higher than observed. The low Nb concentration could be explained if a perovskite-liquid partition coefficient of 13 were assumed for the element. However, if a value of 19 were used (the lower value calculated for the mineral in the southern cluster), the predicted Nb concentration in ZH1 would be lower than observed.

There is a poor agreement between calculated and observed concentrations of the elements Ba, Rb and Y. This is analogous to their behaviour in the southern pipe cluster.

If melilite is included in the mixing calculation (Mix 2, Table 10(b)), agreement between actual and calculated major element compositions for ZH1 are extremely poor, and is particularly bad for Ca. Moreover, predicted concentrations of the incompatible elements (Zr, Sr and Nb) are all appreciably greater than predicted. This suggests that melilite fractionation did not play an important role in modifying bulk rock chemistry in the Gamoep pipes. The low modal melilite proportions in the Gamoep volcanics (Table 5) supports such a conclusion.

Mix 3 in Table 10(b) illustrates predicted and actual concentrations obtained by using samples ZHM3 and ZHC1 (from the Zwartheuwel pipe) and the low pressure phases olivine, titanomagnetite and perovskite in the mixing calculation. Agreement is rather poor, particularly for Al and Ti. If melilite is included in the mix, (Mix 4, Table 10(b)) the agreement is even poorer.

Mix 5 in Table 10(b) illustrates the results obtained using samples (ZHM3 and WK2) from different pipes together with low pressure phases. Agreement between predicted and actual compositions

is once again rather poor. Moreover, the mixing calculation leads to a geologically implausible solution requiring subtraction of olivine, melilite and titanomagnetite, but addition of perovskite to derive composition WK2 from ZHM3. A similar result would be obtained by omitting melilite from the mix. A subjective examination of the Ti-Mg relationship (Fig. 4.5(b)) leads to the same conclusion.

In summary therefore, it would appear that simple addition or subtraction of low pressure phases does not adequately account for the chemical variation observed for the northern pipes.

The best mixes for the northern pipe cluster are obtained using various combinations of high and low pressure mineral phases. Agreement in each case is best if clinopyroxene is used in the calculation, despite the lack of evidence for clinopyroxene phenocrysts.

Mix 6 (Table 10(b)) illustrates the results obtained using the extreme bulk rock compositions for the northern pipe cluster (ZHM3 and ZH1), together with the high pressure phases olivine, clinopyroxene and ilmenite, and the low pressure phase titanomagnetite.

Agreement between actual and predicted concentrations is very good for all major elements except Mn, P, K and Na. It has previously (Section 4.3.2.1) been suggested that Mn would act as a trace component, and that the distribution of the other three elements may have been influenced by loss of a volatile fluid phase, or source area inhomogeneity.

It should be noted that while the other low pressure phases (melilite, perovskite and (hopper) olivine) have been excluded from Mix 6 in Table 10(b), this does not preclude the possibility that these minerals might have had a minor influence on chemical variation in the northern pipes. However, when they are included in

the mixing calculation, geologically implausible solutions result that require addition of some, but subtraction of other phases.

The observed Zr, Nb and Sr concentrations in ZH1 (Mix 6) are all somewhat lower than would be expected by treating them as incompatible elements. These discrepancies may partly reflect partitioning of Zr and Nb into ilmenite and clinopyroxene, and of Sr into the latter phase during fractional crystallization processes. It is difficult to estimate mineral-melt partition coefficients for Zr and Nb because two crystal phases are involved. In order to account for the entire Sr discrepancy, it would be necessary to assume a clinopyroxene-melt partition coefficient of 0.7 for this element. This is much higher than values found for basaltic liquids (up to 0.15), and more similar to those obtained for acidic (highly polymerized) melts (Arth, 1976). This suggests that Sr variation may in part be related to other factors. These may include crystallization of other Ca-rich phases, such as melilite, and possibly also the separation of an immiscible carbonate liquid during magma ascent. At present, however, the behaviour of Sr in the northern pipes is rather poorly understood.

The results for Mix 7 using samples ZHM3 and ZHC1 (from the same pipe) are broadly comparable to those found for Mix 6, although a slightly lower clinopyroxene-liquid partition coefficient (0.47) would be required to account for the Sr concentration in ZHC1.

Mix 8 in Table 10(b) illustrates how samples (ZHM3 and Ho1) from two different pipes could be related by addition or subtraction of a combination of high pressure (olivine, clinopyroxene and ilmenite) and low pressure (melilite and titanomagnetite) phases.

As for earlier mixes, good agreement is found for all major elements apart from Mn, P, K and Na. It should be noted that the high pressure phases dominate the mix proportions, suggesting that low pressure fractionation processes did not play a major role in influencing chemical variation in the northern pipe cluster. Although the low pressure phases perovskite and hopper olivine have been excluded from the mix, this does not necessarily indicate that they did not play a role in modifying bulk rock compositions. However, their influence is likely to have been secondary. Unfortunately, inclusion of too many phases in the mixing calculation results in a geologically implausible solution.

The observed Sr concentrations in Ho1 (Mix 8) are somewhat lower than would be expected assuming incompatible behaviour of the element, though are consistent with the partitioning of Sr into melilite and clinopyroxene during fractionation processes. However, Zr concentrations are considerably lower than would be expected assuming incompatible behaviour. This discrepancy is too large to explain in terms of partitioning of the element into ilmenite and clinopyroxene, and may indicate the influence of a Zr-rich mineral - for example zircon.

Mix 9 (Table 10(b)) indicates that removal of a combination of high and low pressure phases from ZHM3 would lead to a residual composition closely similar to WK2. The greatest discrepancy is found for Ca. This might be related to the influence of Ca-rich minerals such as melilite and perovskite. However, when these minerals are included in the mixing calculations, geologically implausible solutions involving addition and subtraction of pha-

ses result.

In summary, moderately good mixes are feasible for some, though not all Gamoep samples using only low pressure phenocryst phases (hopper olivine, melilite, titanomagnetite and perovskite). As in the case of the southern pipes however, agreement between predicted and actual concentrations for the elements P, Na, K and Mn is generally very poor.

Greatly improved agreement results if both high and low pressure phases are used in the mixing calculations. In such cases, the high pressure phases dominate the mixes. Such mixes allow for a qualitative explanation of the behaviour of Zr, Nb and Sr. It would therefore appear that high pressure crystal fractionation processes played the dominant role in determining bulk rock chemical variation in the Gamoep pipes. Low pressure crystal fractionation and loss of a vapour phase and immiscible carbonite liquid probably played a subordinate role in modifying bulk rock chemistry.

4.3.2.3 *Relationship Between Northern and Southern Pipe Clusters*

Mixing calculations involving high and low pressure mineral phases and an average bulk rock composition for each cluster (Table 3) have been used to determine the possible genetic relationship between the northern and southern group of pipes. Those mixes involving only low pressure phases either result in a very poor correspondence between predicted and actual concentrations for all elements (Table 10(c), Mix 1) or require geologically implausible solutions (Table 10(c), Mix 2). This strongly suggests that compositional variation between the northern and southern pipe

clusters was not determined by low pressure fractionation effects.

The closest correspondence between actual and predicted compositions is obtained using the high pressure phases olivine, clinopyroxene and ilmenite in the mix (Table 10(c), Mix 3). Poorest agreement is for the elements K, Na, P and Na. This suggests that high pressure crystal fractionation combined with secondary processes such as loss of a vapour phase and immiscible carbonate liquid appear to be adequate to explain major element variation between the two pipe clusters.

Behaviour of Zn and Ni is consistent with a fractional crystallization model. If the former element is assumed to be excluded by the crystallizing phases, the predicted average concentration of this element in the Garies pipes (157 ppm Zn) is close to, though slightly higher than, that observed (146 ppm Zn) - Table 10(c), Mix 3. As significant amounts of Zn are incorporated in clinopyroxene and ilmenite, this discrepancy can be readily explained.

Ni variation between the average Gamoep and Garies bulk rock compositions has been modelled in a step-wise fashion analogous to that outlined in Section 4.3.2.1, using the following partition coefficient:

$$D_{\text{Ni}}^{\text{olivine-liquid}} = \frac{124.1}{\text{MgO}} - 2.85$$

$$D_{\text{Ni}}^{\text{ilmenite-liquid}} = D_{\text{Ni}}^{\text{olivine-liquid}}$$

$$D_{\text{Ni}}^{\text{clinopyroxene-liquid}} = 1.5$$

The Gamoep average bulk rock composition was used as a hypothetical parent liquid composition.

The calculated Mg-Ni (liquid) evolution line is illustrated in Fig. 4.10(d). Removal of just over 30% of the high pressure mineral phases from the average Gamoep composition leads to Mg and Ni concentrations extremely close to those of the average Garies composition.

While the behaviour of Zn and Ni support a simple Raleigh fractionation model for relating the northern and southern pipe clusters, a consideration of the behaviour of the incompatible trace elements suggest that somewhat more complex processes were involved. The average concentrations of Zr, Nb and Sr in the southern pipe cluster are grossly higher than would be expected by assuming that these elements are excluded by the crystallizing phases. This discrepancy is increased when allowance is made for the fact that the phases involved will accommodate significant concentrations of these three elements. A number of alternative processes could in principle account for such discrepancies. They include the following:

- (a) Derivation of the parental magma for the southern pipe cluster from a source area enriched in Zr, Nb and Sr relative to the source for the Gamoep magmas.
- (b) Varying degrees of partial melting. The high concentration of incompatible elements in the olivine melilitites relative to MOR basalts (Table 3) suggests derivation of the former by small degrees of partial melting. This is in accord with suggestions made by Kay and Gast (1973). If the primary magma in the Garies pipe cluster were derived by lower degrees of partial melting than was the case for the Gamoep cluster, it is possible that

such respective parental liquids were broadly similar in major element chemistry, but differed strongly in the initial incompatible element concentrations. The higher Zr, Nb and Sr concentrations in the evolved Garies rocks might then be related primarily to differences in the degree of partial melting.

(c) Magma mixing processes might have lead to an enrichment of incompatible elements in a deep seated magma chamber beneath the Garies area prior to eruption. Subsequent low pressure fractionation during magma ascent to the surface would result in further compositional modification.

(d) Zone refining processes (Harris, 1957; and Harris and Middlemost, 1970).

At present, it does not appear to be possible to choose between these different options, although they have strongly divergent implications regarding the processes of magma generation and evolution.

4.3.2.4 *Discussion*

The foregoing analysis suggests that compositional variation in the Gamoep pipes was dominated by high pressure fractionation processes, with low pressure crystal fractionation being relatively unimportant. Secondary processes involving the separation of volatile components and an immiscible carbonate liquid during magma ascent appear to have played an important role in controlling the behaviour of the elements K, Na, P, Ba, Rb and Y. Low pressure crystal fractionation appears to have been the dominant mechanism governing geochemical variation in the southern pipe

cluster, with separation of volatiles and an immiscible carbonate liquid once again leaving a secondary chemical imprint. However, it appears that the parent magma for the southern pipe cluster may have been derived by separation of high pressure phases from a liquid equivalent in major element chemistry to the average Gamoep composition - in other words, by an extension of the fractionation processes controlling magma evolution within the northern pipe cluster.

Field evidence indicates that megacrysts (*i.e.* high pressure phenocrysts) and mantle-derived xenoliths are absent in the southern pipes, though are abundant in some of those in the Gamoep cluster. This relationship is consistent with the model of extensive low pressure fractionation within the former suite of rocks, but only minor low pressure fractionation within the latter. Factors determining the depth at which fractionation would be effective remain speculative, though are likely to be related to the depths at which volatile components and an immiscible carbonate separate from the magma, and the resultant effect on ascent velocities and magma emplacement mechanisms (Sparks *et al.*, 1978).

4.3.2.5 *Experimental Considerations*

Mixing calculations indicate that high pressure crystallization of olivine and clinopyroxene in a ratio of roughly 2:1 would be required to account for the chemical variation found in the Gamoep pipes, and explain the variation in major element chemistry from the average Gamoep to the average Garies composition. However, anhydrous melting studies (O'Hara, 1968) show that high pressure cotectic crystallization of clinopyroxene and olivine

would be dominated by removal of the former phase. At lower pressures, the change in geometry of the primary phase field boundary would be expected to result in even more clinopyroxene-rich extracts during isobaric cotectic crystallization. High pressure cotectic crystallization in CO₂-rich systems would also result in a clinopyroxene-rich extract (Wyllie and Huang, 1976, Fig. 11D).

In order to reconcile experimental and chemical evidence, it appears necessary to assume that magma evolution was not controlled by simple isobaric high pressure fractionation of olivine and clinopyroxene. There is a marked expansion of the olivine primary phase field towards low pressures (O'Hara, 1968), and it is possible that the high proportion of olivine relative to clinopyroxene required to explain the chemical data indicates that polybaric high pressure crystallization occurred. This would imply continuous crystallization during magma ascent, rather than isobaric crystallization as a result of a temporary accumulation in a high pressure magma chamber. Because of the continuous expansion of the olivine primary phase field, olivine proportions would be greater than predicted by a consideration of the geometry of the isobaric phase boundaries.

4.3.2.6 *The Clinopyroxene Dilemma*

The greatest drawback to a model involving extensive high pressure clinopyroxene fractionation in the Namaqualand-Bushmanland volcanics is the absence of clinopyroxene phenocrysts in these rocks. It would appear that such a dilemma can only be resolved by appealing to highly efficient low pressure resorption of this phase.

4.4 PRIMARY MAGMA COMPOSITIONS

Geological opinion is divided on the problem of identifying primary magma compositions. O'Hara (1968) has argued that closed system melting of a garnet lherzolite source will produce a primary picritic liquid, and further, that the range in composition of basic rocks at the earth's surface can be understood largely in terms of a model involving fractional crystallization of such a parent liquid. Other authors (e.g. Green, 1971) suggest that primary liquids of grossly differing compositions will be produced at different depths and differing degrees of partial melting, while recent studies have stressed the importance of volatiles such as water (Green, 1973) and CO₂ (e.g. Brey and Green, 1977; Wyllie and Huang, 1968) in controlling the composition of mantle-derived melts. Against this background of discord, it is necessary to consider whether any of the Namaqualand bulk rock compositions may in fact represent primary magma compositions.

Primitive basic or ultrabasic liquids are generally considered to have been generated in equilibrium with mantle olivine, and if this assumption is valid, strong constraints are placed on the initial Mg/Fe⁺⁺ ratios (Roeder and Emslie, 1970) and Ni concentrations (Gast, 1968; Hart and Davis, 1979; and others) of such liquids.

4.4.1 MAGNESIUM NUMBERS

Kesson (1973) points out that mantle olivine compositions probably fall within the range Fo₈₈-Fo₉₂, and that the magnesium number (100Mg/(Mg + Fe⁺⁺))(atomic proportions) of primary magmas would be expected to vary from 69 to 77 if a value of 0.3 is used

for the Mg-Fe distribution coefficient defined by Roeder and Emslie (1970).

Table 1(k) lists magnesium numbers for the Namaqualand volcanics assuming $\text{Fe}_2\text{O}_3/\text{FeO}$ ratios of 0 and 0.2 respectively.

The magnesium number is greater than 69 for all samples from the northern group of pipes if a $\text{Fe}_2\text{O}_3/\text{FeO}$ ratio of 0.2 is assumed, and varies from 66 to 75 using a $\text{Fe}_2\text{O}_3/\text{FeO}$ ratio of zero. The spread in bulk rock compositions from the Gamoep area may therefore include those with primitive chemistry. However, the Mg-numbers do not uniquely define primitive compositions, or require that any of the Gamoep bulk rock compositions represent primitive liquids. Cumulus enrichment in ferromagnesian phases could also lead to high magnesium numbers.

Using a $\text{Fe}_2\text{O}_3/\text{FeO}$ ratio of 0.2, the calculated magnesium numbers for the southern volcanics are all smaller than those expected of primary magmas in equilibrium with olivines of mantle compositions. This suggests that bulk rock compositions for the southern pipes are not primitive, but have probably been modified by fractional crystallization of a ferromagnesian phase (or phases) unless:

- (a) the $\text{Fe}_2\text{O}_3/\text{FeO}$ ratio of the magma(s) in the source area was appreciably greater than 0.2 or
- (b) these magmas were not in equilibrium with olivine in the source region or
- (c) a value of 0.3 for K_D (as defined by Roeder and Emslie, 1970) was inappropriate to these magmas in the source area or

(d) some combination of these possibilities applied.

In summary, Mg/Fe relationships suggest that the northern pipe cluster might include rocks with primitive chemistry, but that the Garies volcanics are all chemically evolved.

4.4.2 Ni CONSTRAINTS

Average mantle Ni concentrations are likely to be close to 2 000 ppm Ni (Sato, 1977). It should in principle therefore be possible to predict Ni concentrations of mantle-derived liquids with a reasonable degree of confidence. In practise however, interpretation of Ni data is complicated by uncertainty as to whether the element obeys Henry's Law for the range of compositions typical of mantle olivines, and disagreement as to the factors controlling variations in the olivine-liquid partition coefficient for Ni.

Thus, Mysen (1976(b)) argues that Henry's Law will not be valid for Ni-rich olivines, though this is disputed by Hart and Davis (1978) and Drake and Holloway (1978).

Leeman (1973) and Leeman and Lindstrom (1978) suggest that K_{Ni}^{ol-liq} is strongly temperature dependent. Hart and Davis (*op. cit.*) however argue that the apparent temperature control merely reflects liquid composition effects which are more fundamental to determining Ni distribution between olivine and liquid. Based on experimental data for the (iron-free) synthetic system Fo-Ab-An at 1 atmosphere, the latter authors suggest that K_{Ni}^{ol-liq} is related to the MgO content of the liquid by the equation:

$$K_{Ni}^{ol-liq} = \frac{124.1}{MgO(liq)} - 0.9$$

However, data for the partitioning of Ni between olivine and natural (*i.e.* iron-bearing) basic and ultrabasic komatiite compositions (Arndt, 1977) indicate systematically lower values of K_{Ni}^{Mg-liq} than would be expected using the Hart-Davis relationship. H S Smith (oral communication, 1978) has found that if the data of Arndt (*op. cit.*) and Green (1975) are used to calculate Ni partition coefficients, K_{Ni}^{ol-liq} can be related to the MgO content of the liquid by the expression:

$$K_{Ni}^{ol-liq} = \frac{124.1}{MgO(liq)} - 2.85$$

Although such conflicting evidence complicates the interpretation of Ni data for a suite of volcanic rocks, an attempt has been made to use bulk rock Ni variations in the Namaqualand volcanics to identify possible primitive compositions. The following assumptions have been made:

(1) Partial melting in the mantle produces a range of primary magmas with a spread in MgO concentrations. Progressively higher MgO contents reflect higher degrees of partial melting. It has arbitrarily been assumed that a magma with 5% MgO represents a 5% melt in the source area, and that one with 25% MgO represents 15% melt. It is further assumed that the degree of partial melting is linearly related to the MgO concentration of these liquids. Minor errors in these assumptions will not have a marked effect on the melting calculations.

(2) Equilibrium melting occurs and Henry's Law is obeyed. The concentration of Ni in the melt (C_L) will therefore be related to the concentration in the source area (C_0) according to the equa-

tion:

$$\frac{C_L}{C_0} = \frac{1}{D + F(1-P)} \quad \text{(Shaw, 1970)}$$

(See Section 4.4.7.6)

(3) The source area has a bulk Ni concentration of 2 000 ppm (Sun and Hanson, 1975(b)) and the following modal mineralogy:

Olivine	:	60%
Orthopyroxene	:	20%
Clinopyroxene	:	10%
Garnet	:	10%

(4) The partition coefficient for Ni between olivine and coexisting melt can be determined from the relationship:

$$K_{Ni}^{ol-liq} = \frac{124.1}{MgO(liq)} - 2.85 \quad \text{(H S Smith, *op. cit.*)}$$

(5) Ni partition coefficients for melt and the remaining mantle phases are assumed to be constant and have the following values:

$$\begin{aligned} K_{Ni}^{opx-liq} &= 3 \\ K_{Ni}^{cpx-liq} &= 1.5 \\ K_{Ni}^{Gt-liq} &= 1.0 \end{aligned}$$

Errors in the Ni partition coefficients assumed for the above phases will not have a marked influence on the melting calculations, except for very high MgO liquids, which are characterized by low values for K_{Ni}^{ol-liq} .

(6) Modal melting proportions are 60% orthopyroxene and 40% olivine. This assumption can naturally not be realistic. However, the melting calculations are mainly dependent on the value

of the bulk rock-liquid distribution coefficient for elements (such as Ni) that are strongly concentrated in the residue during melting, and would not be markedly influenced by assuming that relatively large proportions of other phases were contributed to the liquid.

Based on the relationship given in assumption 4, values for the Ni partition coefficient for olivine and liquid (K_{Ni}^{ol-liq}) have been calculated for hypothetical melts with different MgO contents. These, together with the assumed partition coefficients for the other mantle mineral phases were then used to calculate a bulk rock-melt distribution coefficient (D) for each of these hypothetical melts, and thus expected Ni concentrations (C_L) for liquids of different MgO contents. The theoretical melting curve is plotted in Fig. 4.10(c) together with the bulk rock Ni-Mg data for the Namaqualand volcanics. (Also shown on the diagram is a second hypothetical melting curve, obtained by assuming that K_{Ni}^{ol-liq} is controlled by the relationship suggested by Hart and Davis (1978).)

For the Gamoep pipes, most bulk rock samples plot close to or on the first theoretical melting curve. It would appear therefore, that allowing for the assumptions made regarding mantle chemistry and mineralogy, the degree of melting, and the factors governing K_{Ni}^{ol-liq} , Ni-Mg variations in the Gamoep pipes are consistent with, though do not prove the possibility that the spread of bulk rock compositions from this cluster may include those with primitive chemistry. However, as in the case of Mg/Fe relationships, a unique primitive composition is not defined. Moreover, if the

Hart-Davis relationship is used to calculate K_{Ni}^{ol-liq} , the theoretical melting curve would be displaced towards Ni-poor concentrations (Fig. 4.10(c)).

For the Garies volcanics, bulk rock compositions are all markedly depleted in Ni relative to both theoretical melting curves. This indicates either that the parental magmas underwent strong fractionation involving a Mg- and Ni-rich phase, or that one or more of the assumptions made for the melting calculations is invalid. (The parent magmas may for example not have been in equilibrium with olivine in the source area.)

In summary therefore, Mg-Fe-Ni relationships strongly point to the Garies volcanics being chemically evolved, but indicate that the Gamoep bulk rock compositions may include those representing primary magma compositions, although do not uniquely define such primitive liquids.

Olivine compositions have bearing on the problem of identifying primary magmas. The hopper olivines that dominate the phenocryst population in the Gamoep rocks are always considerably less ferroitic than might be expected had they crystallized from liquids that had previously equilibrated with olivines of mantle composition. This indicates either that these olivines attained more Fe-rich compositions as a result of continuous re-equilibration with a cooling host magma, that the parent liquid did not equilibrate with olivines of mantle composition, or that the hopper olivine phenocrysts crystallized from relatively evolved liquids. In view of the evidence for relatively rapid magma ascent and crystallization, the latter alternative appears to be the most

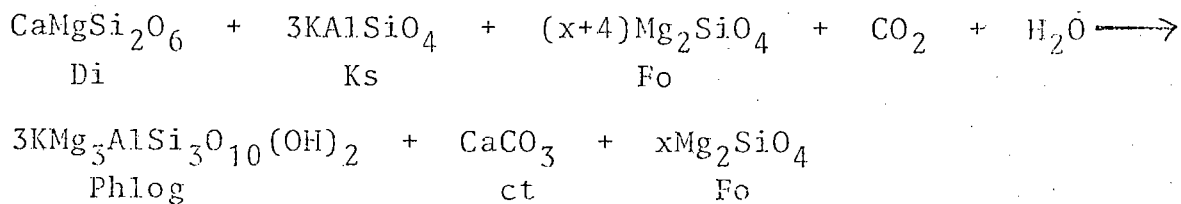
likely. This would imply that none of the Gamoep bulk rock compositions could be regarded as representing a primary magma, and thus that the composition of the parental liquid for the Namaqualand-Bushmanland volcanics remains speculative.

It should be noted moreover, that in view of their marked enrichment in incompatible elements, it is unlikely that any of the Namaqualand-Bushmanland bulk rock compositions represent liquids derived from a four-phase garnet lherzolite source. Thus if they do indeed include primary magma compositions, it would be necessary to invoke a source enriched in incompatible elements and volatiles such as CO_2 , possibly as a result of upper mantle metasomatism such as is envisaged by Erlank (1976) and Aoki (1976). If so however, such metasomatism could not have occurred long before the eruption of the Namaqualand-Bushmanland volcanics, for these rocks are not markedly enriched in radiogenic strontium (available data show a range in $^{87}\text{Sr}/^{86}\text{Sr}$ from 0.70338 ± 6 to 0.70408 ± 4 ; Table 1(j)), as would be expected if derived from a source that was enriched in Rb over an extended period prior to magma generation. This would raise the intriguing possibility that mantle metasomatism was responsible for triggering volcanic activity.

5. RELATIONSHIP BETWEEN OLIVINE MELILITITES AND KIMBERLITES

5.1 GENERAL

Numerous authors have recognized the close kinship between kimberlites and olivine melilitites, and have speculated on the genetic link between the two rock types. Taljaard (1936) proposed that kimberlites might be simply hydrothermally altered olivine melilitites. Holmes (1936) suggested that kimberlites might be regarded as being composite mixtures of a magmatic component, similar in chemistry to olivine melilitites, volatiles such as CO₂ and H₂O, and xenolithic peridotites. This latter view has received recent strong support from Yoder (1975), who suggested that *"a magma having melilitite affinities could be transformed into kimberlite with the aid of suitable volatiles. Loss of volatiles in transit or crystallization at low pressure would yield a melilitite-bearing assemblage in the upper crust"*. Yoder (op. cit.) suggested that at low pressures, reactions such as:



might control the groundmass mineral assemblage of kimberlites.

These views have not been generally accepted, partly no doubt because olivine melilitites and kimberlites appear superficially to be grossly dissimilar. The former are typically hard, fresh basaltic rocks whereas kimberlites are characteristically serpentinized to varying degrees. In addition,

the groundmass mineral assemblages of the two rock types are dissimilar and kimberlites always have appreciable amounts of late-crystallizing carbonate. This phase is all but absent in the olivine melilitites, where it has only been found as inclusions in olivines. This is reflected in their low bulk rock CO_2 contents (typically less than 0.3% CO_2). There are nevertheless many striking mineralogical similarities between the two rock types. Moreover, although mineral chemistry data for kimberlites are surprisingly scarce, the limited number of published analyses for olivines, ilmenites and megacrysts in these rocks suggest that they may have followed a path of physico-chemical evolution which has strong similarities to that proposed for the olivine melilitites. It is instructive to consider the similarities and differences in the mineralogy and mineral chemistry of olivine melilitites and kimberlites, and the significance of the differences.

5.2 MINERALOGICAL SIMILARITIES BETWEEN OLIVINE MELILITITES AND KIMBERLITES

5.2.1 OLIVINE

Olivine is present as a dominant phase in both olivine melilitites and kimberlites. Euhedral and subhedral olivines, which are presumed to be cognate phenocrysts, are common in relatively unaltered kimberlites (Boyd and Clement, 1977). These phenocrysts lack skeletal habit, and in this respect differ from the idiomorphic olivines in many of the Mg-poor olivine melilitites. However skeletal habit is poorly developed in the most Mg-rich olivine melilitites, which are in

turn depleted in Mg relative to kimberlites. Skeletal habit may therefore be related either directly or indirectly to bulk rock compositions.

Data published by Mitchell (1973) show that cores of euhedral olivines from the Wesselton kimberlite (which he terms "groundmass" olivines), define a trend of decreasing Mg/Fe and Ni and that the margins of such olivines are often enriched in Mg relative to the cores. Marginal reversed zoning in kimberlite olivines has also been reported by Dawson and Hawthorne (1973), Emeleus and Andrews (1975) and Boyd and Clement (1977). These patterns of chemical variation are closely analogous to those defined by the hopper olivine populations in the olivine melilitites.

Mitchell (1973) notes that the large rounded olivines in the Wesselton kimberlite (which he considers to be phenocrysts) are commonly more iron-rich than the associated idiomorphic "groundmass" olivines, show a wide range in Ni content, and do not appear to define any coherent pattern of Mg/Fe variation. Nixon and Boyd (1973(b)) record a wide range in composition for the large rounded olivines from the Liphobong kimberlite, while Emeleus and Andrews (1975) have established a range of Fo_{76} to Fo_{88} in olivines from some Greenland intrusions. The latter authors' data indicate that the more iron-rich individuals show reversed zonation towards more Mg-rich compositions. It is suggested that such zonation patterns are analogous to those defined by the unusual hoppers in the Namaqualand olivine melilitites.

The limited data available make it difficult to identify HILN olivines in kimberlites. By definition, such olivines are relatively iron-rich and Ni-poor compared to the idiomorphic olivines in the same rock. In kimberlites they may be represented by the most fayalitic olivines in the megacryst suite (e.g. Fo₇₈, 0.09% Ni, Gurney *et al.*, 1977). Nixon and Boyd (1973(b), Table 45) present analyses of idiomorphic and large rounded olivines from the Lihobong kimberlite. The former tend to be highly fosteritic (up to Fo₉₅) while the latter show a range in composition and include a relatively iron-rich and Ni-depleted individual (Fo₈₇, 0.01% Ni) which could be a HILN olivine. Differences in composition of inferred HILN olivines in kimberlites and those in the olivine melilitites may reflect:

- (i) The small number of published analyses of olivines from kimberlites.
- (ii) The relatively high bulk rock nickel and magnesium contents of kimberlites, which might result in the crystallization of HILN olivines that are enriched in these elements relative to HILN olivines in the olivine melilitites.
- (iii) Higher oxygen activities in kimberlites than those attained in the olivine melilitites.

Gurney *et al.* (1977) report a range in composition of Fo₈₈ to Fo₇₈ for olivine megacrysts from the Monastery kimberlite. Their data indicate a compositional gap from Fo₈₂ to Fo₈₄, which may reflect either inadequate sampling, or the presence

of two distinct olivine megacryst populations. The most fayalitic megacrysts are iron-rich and Ni-poor (Fo₇₈, 0.09% Ni) relative to compositions from several kimberlites (Mitchell, 1973; Emeleus and Andrews, 1975; Boyd and Clement, 1977). There are no published data for idiomorphic olivines from the Monastery kimberlite. However, a bulk rock analysis reported by Gurney *et al.* (1973) gives an Mg/(Mg + Fe) ratio of 79.3 (Fe = total iron). Olivines in equilibrium with liquids of analogous composition would normally be expected to be highly fosteritic, and therefore more magnesium-rich than the iron-rich megacrysts. There is thus evidence to suggest that iron-rich kimberlitic megacrysts will prove to have a compositional relationship to idiomorphic olivines analogous to that which has been established for megacrysts and hoppers from the Zwartheuwel olivine melilitite.

The identification of olivine populations in kimberlites that are closely analogous to those defined in olivine melilitites suggests that the two rock types followed closely similar paths of physico-chemical evolution.

5.2.2 PICROILMENITE

Picroilmenites with up to 12% MgO, although rare, are found in many of the olivine melilitites. Their compositions fall within the range reported for kimberlitic picroilmenites (Haggerty, 1973, 1975), although Cr contents in the latter are sometimes (though not invariably) grossly higher than in those from the olivine melilitites which have been analysed.

The model proposed for the evolution of the olivine melilitite magmas offers a possible explanation for unusual chemical features found in some kimberlite ilmenites. Haggerty (1975) has described ilmenites from the Lihobong kimberlite that show reversed zoning with respect to Mg/Fe. These chemical trends may reflect changing partition coefficients associated with loss of CO₂ and depolymerization of the magma during ascent (see Section 3.2.2.4). The Lihobong ilmenites are characterized by complex reaction rims. The reaction rim sequence is magnesioferrite spinel - secondary ilmenite - Ti-ferrian pleonaste - perovskite, and is ascribed by Haggerty to changing oxygen activity of the magma. A more detailed explanation is feasible if oxygen activities on the carbon saturation surface are assumed to be appropriate to kimberlites. It is then possible to envisage a complex series of intersections of the carbon-gas and various solid-solid buffers as a result of changing temperature and pressure during the final emplacement of the magma. Fig. 5.1 illustrates a possible magma path (A-B-C) through pressure-temperature and f_{O₂} space. At 2 Kb and 900°C (A) the carbon-gas buffer lies above the wustite-magnetite curve. An essentially isothermal pressure decrease from 2 Kb to 1 bar related to the final emplacement of the kimberlite (A to B) would cause the carbon-gas buffer to fall below the wustite-iron curve. With subsequent isobaric cooling, the C-CO-CO₂ buffer would rise above the iron-wustite and wustite-magnetite buffers (B to C). Such an emplacement sequence might result in the formation of a relatively oxidized phase (magnesian ferrite spinel) at A, a re-

latively reduced phase (secondary ilmenite) at B and a second more oxidized phase (Ti-ferrian pleonaste) at C. Note, however, that although an oxygen activity approximating the carbon-gas buffer has been assumed for the Liqhobong kimberlite, these principles could be applied to other external gas buffers.

5.2.3 *PEROVSKITE AND TITANOMAGNETITE*

Ubiquitous microphenocrysts of perovskite and titanomagnetite are characteristic of both olivine melilitites and kimberlites.

5.2.4 *MANTLE-DERIVED XENOLITHS*

Rare garnet lherzolites have been found in one of the Namaqualand olivine melilitite pipes, and chrome-rich garnet and clinopyroxene are commonly recovered from borehole concentrates from many of the Namaqualand pseudo-kimberlites, including a number which closely resemble hydrothermally altered olivine melilitites. These accidental inclusions are similar to those reported from kimberlites.

5.3 *MINERALOGICAL DIFFERENCES BETWEEN OLIVINE MELILITITES AND KIMBERLITES*

- (a) Diamonds have never been reported from olivine melilitites.
- (b) Ultramafic xenoliths and associated xenocrysts are far more common in kimberlites than in olivine melilitites. Pressures of equilibration of xenoliths in the former

pipes (e.g. Boyd and Nixon, 1975) are generally higher than for those in the olivine melilitites (Moore, 1973).

- (c) Melilite is invariably absent in kimberlites (Clement *et al.*, 1977).
- (d) There are marked differences between the groundmass assemblages of olivine melilitites and kimberlites. Clinopyroxene and nepheline are invariably the dominant late-stage minerals in the former rocks, whereas in kimberlites, the groundmass normally consists of varying proportions of the minerals phlogopite, calcite, serpentine, diopside, montechellite and apatite (Clement *et al.*, 1977).

5.4 FACTORS DETERMINING THE MINERALOGICAL DIFFERENCES BETWEEN KIMBERLITES AND OLIVINE MELILITITES

Diamonds have been found in exotic eclogite and peridotite inclusions in kimberlites (Hatton and Gurney, 1977; Dawson and Smith, 1975), and it is possible that many if not all diamonds are derived from disaggregated mantle material. The absence of a carbon phase in the olivine melilitites may therefore be partly related to the rarity of ultrabasic xenoliths in these rocks, which in turn may be largely determined by the interaction of magma ascent rates, settling velocities and rates of resorption of unstable phases, and not of genetic importance. Thus it would appear to be unwise to attach great significance to the absence of diamond in the olivine melilitites until the factors governing the occurrence of the phase

in kimberlites are fully understood. The most important mineralogical differences between olivine melilitites and kimberlites would therefore appear to lie in their contrasting groundmass assemblages and the absence of melilite in the latter rocks. In other words, the two rock types appear to have followed rather similar evolutionary paths until the relatively late stages of crystallization. It is therefore necessary to consider the factors controlling the differences in the late-crystallizing minerals in olivine melilitites and kimberlites.

It has been pointed out (Section 3.2.1) that the presence of melilite in the Namaqualand volcanics is related to both the emplacement and cooling history of the magma, and to the relative abundance of alkalis (Velde and Yoder, 1975; Yoder and Velde, 1975). Moreover, the stability field of melilite contracts in the presence of H_2O and CO_2 (Yoder, 1973). The absence of this mineral in kimberlites may therefore be related to the abundance of volatiles in these magmas, and possibly also rapid ascent rates within the melilite stability field.

The contrasting groundmass assemblages of kimberlites and olivine melilitites can also be explained in terms of the different volatile contents of the respective magmas at the time of emplacement in the upper crust. Crystallization of phlogopite in kimberlites could be ascribed to late-stage reactions such as those proposed by Yoder (1975). Stability of the mineral would probably be enhanced by the high K/Na ratios

(which reflect relatively low Na concentrations) typical of these rocks. Such chemical characteristics may in part control the absence of nepheline in kimberlites. Their significance is discussed ahead.

Serpentine is a common groundmass mineral in kimberlite (Clement *et al.*, 1977). The phase is unstable at elevated temperatures. However, if kimberlite emplacement temperatures were as low as those suggested by McFadden and Jones (1977) - roughly 350°C - serpentine may have crystallized as a primary late-stage magnesian phase, perhaps at the expense of clinopyroxene, which is a ubiquitous groundmass mineral in olivine melilitites.

The presence of calcite in kimberlites probably reflects a high CO₂ content in the magma at the time of emplacement. The mineral may therefore have crystallized as a primary phase, either directly from the kimberlite magma, or from a late-stage immiscible carbonate liquid (Clement, 1975). The absence of groundmass carbonates in the olivine melilitites is ascribed to the loss of CO₂ and an immiscible carbonate fraction during magma ascent. The rounded carbonate inclusions and the fluid inclusions in the hopper olivines are considered to record the loss of such components. Carbonate dykes are found to the east of the Gamoep pipe cluster (H Jenner-Clarke, oral communication) and are associated with the Zandkopsdrift pipe (Garies cluster). These may be the field expression of carbonate liquids associated with the olivine melilitites.

Loss of CO₂ and an immiscible liquid from silicate melts pro-

bably reflects the decreasing solubility of the former components in the latter phase with decreasing pressure (Mysen *et al.*, 1975). The reason for the efficient separation of CO₂ and an immiscible carbonate liquid from olivine melilitites but not kimberlites is unclear but may have been controlled by several complementary factors: Holloway and Eggler (1976) suggest that the solubility of CO₂ in silicate melts should increase in this order of network modifying cations: Mg, Ca, Na, K. Although kimberlites are in general depleted in Ca relative to olivine melilitites, they are markedly enriched in Mg. It is possible that as a result of the interaction of these chemical characteristics, CO₂ solubility in the former magmas would be higher than in the latter, other factors being equal. Saturation with respect to this component might thus be expected to occur at shallower depths in kimberlites than in olivine melilitites. At such shallow depths, rapid expansion and exsolution of gases dissolved in the magma might lead to a rapid final emplacement, with insufficient time for the complete separation of the immiscible silicate and carbonate phases. The volume of gases present in kimberlite magmas at low pressures would probably be enhanced by the decomposition of the carbonate anion to CO₂ associated with the depolymerization of the magma (Section 3.2.2.4). A consequence of the rapid expansion of this and associated gases would be a strong, adiabatic cooling, which probably determined the low emplacement temperatures typical of kimberlites.

~~There are two further factors which may have lead to kimber-~~

A further factor might have influenced the depth at which an immiscible carbonate and CO₂-rich vapour began to separate from kimberlite magmas:

Mysen *et al.* (1975) demonstrates that CO₂ solubilities in silicate melts increase with increasing temperature. Because kimberlites are in general enriched in MgO relative to olivine melilitites, they would be expected to have higher liquidus temperatures than the latter magmas. Such higher temperatures would counter the effect of decreasing CO₂ solubility with pressure until relatively shallow depths at which rapid degassing would occur.

Field evidence (Section 2) indicates that the emplacement of olivine melilitite in the upper crust was probably accompanied by a convective downflow of magma at the pipe margins. A relatively gentle magma overturn during ascent might have provided a very efficient mechanism for kneading an immiscible carbonate phase from the complementary silicate liquid.

In summary therefore, there are many strong mineralogical similarities between kimberlites and olivine melilitites. The principal mineralogical differences are in the contrasting groundmass assemblages of the two rock types, and these can be explained in terms of the higher volatile contents of the former magmas at the time of crystallization at upper crustal levels. This in turn appears to relate to the efficient loss of volatiles and an immiscible carbonate liquid from olivine melilitites but inefficient separation of these components from kimberlites. This suggests that the evolutionary paths of the two respective magmas were closely similar, and differed chiefly in their late-stage history.

5.5 CHEMICAL RELATIONSHIP BETWEEN OLIVINE MELILITITES AND SOUTH AFRICAN KIMBERLITES

5.5.1 *RELATIONSHIP BETWEEN THE NAMAQUALAND-BUSHMANLAND OLIVINE MELILITITES AND ASSOCIATED "PSEUDO-KIMBERLITES"*

Relative to the Namaqualand-Bushmanland olivine melilitites, the associated "pseudo-kimberlites" are depleted in Fe, Ca, Rb and Ba (Figs. 4.3(a), 4.4(a), 4.21 and 4.23) and enriched in SiO_2 and H_2O (Fig. 4.2(a)). The concentration ranges for the remaining elements analysed partially overlap those established for the olivine melilitites, although the compositional fields for the two rock types are slightly separated in many binary plots. An exception to this rule is found for the Zr-Nb compositional fields (Fig. 4.27) which show complete overlap for the two rock types. Zr and Nb are generally considered to be immobile during alteration processes (Pearce and Cann, 1971), and the overlap of the fields of the two rock types on the Zr-Nb binary plot supports the field interpretation of the "pseudo-kimberlites" being merely altered equivalents of the olivine melilitites. The separate compositional fields for the two rock types as defined by many other element pairs might therefore be primarily related to alteration processes. However, to establish such a relationship with any confidence, it would be necessary to carry out a detailed study of the chemical variation of the "pseudo-kimberlites" and of the effects of alteration to the olivine melilitites. Suitable samples for such a study were not available to the author. Nevertheless, whatever the kinship of these rocks, it is clear that the "pseudo-kimberlites" analysed are comparatively MgO-poor, and in this respect differ markedly in chemistry from

typical kimberlites.

5.5.2 BULK ROCK CHEMISTRY OF SOUTH AFRICAN DIAMOND-BEARING KIMBERLITES

Figs. 4.n(b) illustrate bulk rock chemical data for a suite of South African diamondiferous kimberlites, and their relationship to the Namaqualand-Bushmanland olivine melilitites. Chemical data for the former rocks are from unpublished analyses kindly made available by Drs C R Clement (De Beers Consolidated Mines) and J J Gurney (University of Cape Town). Because the model for the olivine melilitites appeals to the separation of volatiles and an immiscible carbonate liquid during ascent, the kimberlite data have been recalculated on an anhydrous, calcite-free basis. The elements Sr, Zr, Nb, Ca, P and Y all show a strong positive correlation with bulk rock CO₂ contents in kimberlites (Figs. 4.43 - 4.46). This indicates that the chemical behaviour of these elements will be strongly controlled by the proportion of carbonate present in the rock. Consequently, in order to attempt to monitor the effects of processes such as crystal fractionation, only those samples with less than 4% CO₂ have been used in the plots. Such a "window" was also aimed at eliminating highly altered specimens.

Because detailed mineralogical data for the kimberlite suite were not available to the author, and as the relatively Mg-poor, Si-rich samples appear to be contaminated by crustal material (C R Clement, oral communication), it is felt that only a qualitative discussion of the kimberlite data is warranted at this stage.

Consideration of the chemical variation of the major elements

Al_2O_3 , SiO_2 , Fe_2O_3 , CaO and TiO_2 (Figs. 4.1(a)- 4.5(a)) suggests that kimberlite bulk rock compositions can be related essentially by addition or subtraction of varying proportions of olivine and ilmenite. This is indeed not surprising. Olivine is almost invariably the dominant phenocryst phase in kimberlites while ilmenite is commonly a dominant component of kimberlite concentrates. However, it does not follow that simple Raleigh-type fractionation occurred. Most kimberlite pipes are composite intrusions, with several generations of kimberlite present. C R Clement (oral communication) notes that in cases where it has been possible to establish a relative age sequence for these different intrusions, there does not appear to be any systematic chemical variation with time, as might be expected if they tapped a common magma chamber containing a liquid being modified by progressive crystal fractionation. A possible explanation for such a relationship - or rather lack of one - is that a mechanical mixing of solid phases and residual fluids occurred during turbulent emplacement of the kimberlite magma, and that bulk rock chemical variation was dominantly determined by such a mixing process.

In contrast to the fairly coherent behaviour of the majority of the major elements relative to MgO , the elements Zr, Nb, Sr and P (Figs. 4.18(b), 4.19(b), 4.25(b) and 4.6(b)) and Y, Rb, Ba, K and Na do not display well-defined chemical variation patterns. This probably reflects the strong influence of a carbonate component on the distribution of the first five elements, and the effects of separation of volatile components on the latter four. It is also possible that fractionation of minor phases such as zircon, which occurs as discrete megacrysts, may have strongly

influenced chemical variation of minor elements such as Zr.

5.5.3 RELATIONSHIP BETWEEN DIAMOND-BEARING SOUTH AFRICAN KIMBERLITES AND OLIVINE MELILITITES

Relative to the Namaqualand-Bushmanland olivine melilitites, kimberlites are in general enriched in MgO and SiO₂ but depleted in Al₂O₃, Fe₂O₃, CaO and TiO₂. When the latter four elements are plotted against MgO, the field of kimberlite compositions straddles the continuation of the overall chemical trends defined by the olivine melilitites, suggesting a systematic relationship between the two rock types.

The kimberlites are in general depleted in the incompatible elements P₂O₅, Zr, Nb, Sr and Y relative to the Garies olivine melilitites and the concentration of these elements in many of the kimberlite samples correspond to the low end of the concentration range for the Gamoep volcanics (Figs. 4.6(b), 4.18(b), 4.19(b) and 4.25(b); Y data not illustrated). These elements all show a strong positive correlation with CO₂ (i.e. carbonate) concentrations in kimberlites. The model for the evolution of the olivine melilitites involves the separation of an immiscible carbonate liquid during ascent. This process would therefore be expected to result in a relative depletion of these elements in the complementary silicate liquid. Conversely, it seems likely that the original homogeneous CO₂-rich parent olivine melilitite magmas may have been enriched in these elements relative to bulk rock compositions of the olivine melilitites, and possibly also relative to the kimberlite bulk rock compositions. If so, this would imply an overall decrease in their concentration with in-

creasing Mg bulk rock content - *i.e.* from compositions represented by the Garies olivine melilitite protomagmas through those represented by the Gamoep protomagmas to kimberlites.

The Mg-rich character of kimberlites suggests that they may be more primitive rock types than olivine melilitites. This is supported by the presence of abundant highly fosteritic (up to Fo₉₄) olivines in many kimberlites (Mitchell, Emeleus and Andrews, 1975; Boyd and Clement, 1977). This indicates that the liquids from which such olivines crystallized may originally have been in equilibrium with mantle olivine - in other words that they were relatively unfractionated. In contrast, the more iron-rich (up to Fo₈₆) idiomorphic olivines characteristic of even the most Mg-rich olivine melilitites suggest crystallization from a more evolved liquid. However, in the absence of petrological and mineral chemistry data for kimberlites, it is not possible to formulate a detailed model to account for the chemical differences between these rocks and olivine melilitites. Thus, it is not yet certain whether the latter could be derived from kimberlites by a process of crystal fractionation combined with loss of an immiscible carbonate and volatile components or whether factors such as the depth and degree of partial melting played an important role in determining the chemical differences between the two magma types.

It is however possible to speculate on the significance of one striking chemical difference between kimberlites and olivine melilitites - that of the relatively low Na contents and consequently higher K/Na ratios in the former rock types. There are several

alternative explanations that might account for such chemical differences:

The low Na contents in kimberlites could be interpreted in terms of derivation from a source depleted in Na, or one which contained a Na-rich mineral that remained stable during melting, although such a phase is difficult to envisage. An alternative, and favoured explanation for the low Na contents of kimberlites is that the element has been selectively removed from these magmas in a vapour phase during ascent to the surface. Van Groos and Wyllie (1973) point out that in equilibria involving silicate and carbonate liquids and a vapour, Na is strongly partitioned into the latter phase. If saturation with respect to CO₂ occurred at a shallower depth in kimberlite magmas relative to olivine melilitite liquids, it is plausible that a greater proportion of this component would be lost as vapour in the former liquids, while in the latter, greater proportions would be lost as an immiscible carbonate liquid.

If Na has been selectively removed from kimberlites by a vapour transport mechanism, it would be expected that the wall rocks would show evidence for this process in the form of fenitization effects analogous to those associated with many alkaline igneous complexes. However, as kimberlites are small bodies in comparison to such complexes, the fenitized aureole would be expected to be restricted. Moreover, kimberlite temperatures in the upper crust tend to be low (McFadden and Jones, 1977), presumably as a result of the cooling associated with rapid expansion of volcanic gases at upper crustal levels. As a result, temperatures in the wall rocks may have been too low for efficient fenitization pro-

cesses to take place.

5.5.4 HIGH MG CONTENTS OF KIMBERLITES

A primary parent magma for kimberlites has not been unequivocally identified, although it is likely to have been Mg-rich, and may be represented by a roughly intermediate kimberlite composition. This would imply MgO contents of about 30%. The high concentrations of incompatible elements in these rocks indicates that they cannot have been derived by high degrees of partial melting, as has been suggested for ultramafic komatiites with equivalent MgO concentrations. It is therefore necessary to consider the factors controlling the Mg-rich character of kimberlites. They may include the following:

- 1) The olivine-liquid partition coefficients may be higher for carbonate-rich magmas than for CO₂-free magmas of equivalent composition (Section 3). This would in turn imply that carbonate-rich liquids generated in equilibrium with mantle olivines would be more Mg-rich than carbonate-free magmas generated in equilibrium with olivines of identical composition.

- 2) Kimberlites are enriched in Fe and Ti relative to ultrabasic rocks such as komatiites with equivalent MgO contents. This may indicate that the former volcanics were derived from a source containing a Fe-Ti bearing mineral (e.g. ilmenite) which broke down during the early stages of melting to produce a relatively Fe-rich initial liquid. In order to remain in equilibrium with mantle olivine, such a liquid would also have to be Mg-rich.

It should be noted that the two alternatives presented are not

mutually exclusive.

In summary, it is suggested that the mineralogical differences between olivine melilitites and kimberlites can be explained largely in terms of efficient separation of volatiles and an immiscible carbonate fluid from the former magmas, but inefficient separation of these components from the kimberlites. Factors controlling the chemical differences between the two rock types are not yet established, but the chemistry of the kimberlites indicates that they are more primitive rock types.

The depth at which volatiles are lost and a carbonate liquid begins to separate from the respective magmas will be dependent on several factors. The following (discussed in detail in Section 5.4) are probably the most important:

- (i) Compositional effects, in particular the MgO content of the liquid.
- (ii) Magma temperature, as carbonate solubility in silicate liquids shows a positive temperature dependence (Mysen *et al.*, 1975).

5.5.5 TECTONIC IMPLICATIONS OF CHEMICAL DIFFERENCES BETWEEN OLIVINE MELILITITES AND KIMBERLITES

Diamondiferous kimberlites are confined to the central parts of the craton in southern Africa (Dawson, 1971), whilst olivine melilitites occur closer to the continental margin. The chemical characteristics of kimberlites, Gamoep olivine melilitites and Garies olivine melilitites indicate a progressive decrease in bulk rock MgO contents of these alkaline volcanics from the interior of the craton outwards. The significance of this pattern is

uncertain, though may indicate a relationship between lithospheric thickness and the depth at which magmas are generated, or alternatively, the degree to which they evolve by crystal fractionation during ascent.

6. TECTONIC FRAMEWORK OF POST-GONDWANALAND ALKALINE VOLCANISM IN SOUTHERN AFRICA

6.1 GENERAL

Plate tectonics concepts provide a unifying framework for interpreting volcanic activity at plate margins. However, the factors controlling intraplate volcanism, which is frequently alkaline in nature, remain poorly understood. In this section, it is intended to draw attention to apparent relationships which may have bearing on the origin of alkaline igneous rocks in southern Africa. These are not yet unequivocally proven by available data, but are felt to be sufficiently intriguing to stimulate further investigation. Thus while suggestions are made regarding the significance of these apparent relationships, they should at this stage still be regarded as speculative.

6.2 AGES OF ALKALINE VOLCANICS IN SOUTHERN AFRICA

Moore (1976) has suggested that there have been four distinct episodes of alkaline igneous activity in southern Africa since the opening of the proto-Atlantic some 127 m.y. ago. The oldest of these is represented by early Cretaceous sub-volcanic complexes which define alkaline igneous lineaments in the west of the country, and have a range in ages spanning the time of separation of Africa and South America. The three younger events (Turonian, late Cretaceous-early Tertiary and Oligocene) are represented by a variety of volcanic pipes, including kimberlites, olivine melilitites, olivine nepheli-

nites, phonolites and trachytes, pointing to a distinctly different style of alkaline igneous activity compared to that represented by the early Cretaceous sub-volcanic complexes.

Many new radiometric age determinations for alkaline rocks in southern Africa are now available, and are summarized together with earlier data in Table 12. There are an appreciable number of kimberlite ages, and several localities have been dated using different techniques. For the Monastery pipe, the agreement between the Rb/Sr, $^{206}\text{Pb}/^{238}\text{U}$ and K/Ar ages is excellent and indicate pipe emplacement at 90 m.y. (Turonian). There is similarly a close agreement in Rb/Sr and $^{206}\text{Pb}/^{238}\text{U}$ ages for the De Beers pipe, which is roughly coeval with the Monastery kimberlite. However, Rb/Sr and $^{206}\text{Pb}/^{238}\text{U}$ ages for the Roberts Victor pipe are grossly disparate (127 m.y. and 92.2 m.y. respectively). Moreover, three zircons from the Bultfontein kimberlite give a range in ages from 81.7 m.y. to 91.2 m.y. Such disagreement severely complicates the interpretation of the radiometric data. Several alternative explanations could be advanced to account for the different ages obtained for the same pipe, and it is not yet possible to establish which is applicable in each case. They include the following:

- (a) Different ages for the same pipe may represent those of different intrusions over a considerable period of time. Many kimberlites are characterized by multiple intrusions (Hawthorne, 1975).

- (b) Post emplacement alteration may result in a spurious age.
- (c) The material analysed may have been exotic and in isotopic dis-equilibrium in the source area. The age would therefore not record the time of pipe emplacement.
- (d) Analytical error. This would lead to an error of less than 100 000 years for the $^{206}\text{Pb}/^{238}\text{U}$ zircon ages (Davis, 1977) but probably 2-4 m.y. for the K/Ar and Rb/Sr determinations.

The zircons have been interpreted as being part of the discrete nodule association (Davis, op. cit.), which Gurney *et al.* (1977) imply are cognate kimberlite minerals. If such interpretations are correct, then the zircon ages would probably be the most reliable estimates of the time of pipe emplacement. However, in view of the uncertainties involved, the significance of many of the ages listed in Table 12 must be considered equivocal.

Figure 6.1 is a histogram of all available radiometric ages. They show a considerable spread, but define four maxima which correspond to those inferred by Moore (1976). Prof. R V Dingle (oral communication, 1979) points out that these data suggest a periodicity of roughly 30 m.y. for the apparent peaks in volcanic intensity on the subcontinent. Whether the spread in ages between the maxima is an artifact of errors associated with the different techniques used or reflects continuous volcanism is at present uncertain.

Marsh (1973) has interpreted the lineaments defined by the early Cretaceous sub-volcanic complexes as representing lines of weakness which lie on the same small circles about the Africa-South America plate rotation pole as major oceanic transform faults. He suggested that igneous activity was triggered by the build-up of stress along such lines of weakness as a result of the break-up of Gondwanaland.

In contrast to the prominent lineaments defined by the early Cretaceous complexes, the later kimberlites and other alkaline pipes have a wide and irregular distribution across the sub-continent, indicating that the origin of these latter volcanics is not readily explained in terms of the model proposed by Marsh. Moore (1976) has suggested that the three younger periods of volcanic activity all show a close temporal relationship to major breaks in the offshore sedimentary record. Such breaks were interpreted as representing major marine regressions related to epeirogenic upwarping of the sub-continent, and it was suggested that volcanism was triggered by sub-lithospheric stresses related to such upwarping. It is therefore instructive to consider the pattern of such upwarping.

6.3 EPEIROGENIC UPWARP PATTERNS

Du Toit (1933) has proposed a rather complex pattern of epeirogenic warping of the sub-continent during the Tertiary and Pleistocene. The elevation of Cretaceous, Tertiary and Pleistocene marine sediments and the gentle seaward dip of

some of these sediments was ascribed to this warping. Fig. 6.2 shows the loci of his inferred axes of maximum uplift. With the exception of the southwest portion of the Griqualand-Transvaal axis, they follow major river divides. Du Toit (op. cit.) regarded the Orange River as being an antecedent course, and interpreted a pronounced steepening in river gradient as marking the point of intersection with the Griqualand-Transvaal axis. Large pans to the south of this axis on the north-draining Hartebeest River were ascribed to the ponding effect of the upwarping.

Recent data from Carrington and Kensley (1969) and Tankard (1976) show that Pleistocene raised beaches on the Namaqualand coast to the west of Garies occur at markedly higher levels than elsewhere along the west coast. These unusually high level raised beaches lie on the coastward extension of the Griqualand-Transvaal axis, and their elevation may therefore be related to recent uplift along that axis. This supports Du Toit's observation that upwarping took place during the Pleistocene, and is probably still active.

King (1963) proposed that the escarpment river divide also represents a line of epeirogenic uplift. This axis is shown by the dashed line in Fig. 6.2. The west coast escarpment axis runs in a roughly north-south direction, and thus across the prominent NE-SW lines of uplift proposed by Du Toit. This suggests that the pattern of uplift may have varied during the Tertiary and the Pleistocene. River drainage basins in southern Africa are separated by a series of regularly

spaced and roughly concentric river divides (Fig. 6.3). The significance of this pattern is not clear, particularly as drainage divides in the interior of the sub-continent are poorly defined, but, by analogy with Du Toit's interpretation, may be related to vertical movements of the sub-continent. Such movements must however have been of pre-Pleistocene age, and may therefore have been related to uplift during the Tertiary and possibly also the Cretaceous period. Episodes of alkaline volcanism may therefore have been associated with uplift along such hypothetical earlier axes. Fig. 6.4 shows a possible distribution of Cretaceous and Tertiary upwarp axes. They differ from those proposed by Du Toit and King in the following aspects:

- (i) A northern (Okavango) axis is identified, separating poorly defined drainage basins in the north of Botswana and Namibia.
- (ii) The Rhodesian axis is extended in an E-W direction (dashed line) and then swings to the NW.
- (iii) The Transvaal axis swings to the NW (dashed line) rather than extending SW towards the west coast. The NW extension of this axis crosses the palaeocourse of the Molopo River, which previously drained southwards into the Orange River. The Molopo now terminates in a pan just to the north of the proposed NW extension of Griqualand-Transvaal axis, and it is possible that the original drainage pattern was disrupted by the upwarping (see Fig. 6.3).

- (iv) The escarpment axis is arbitrarily placed slightly coastward of the escarpment river divide (shown by the short-dash line) on the assumption that there has been some degree of scarp retreat subsequent to uplift.

The proposed distribution of Cretaceous-early Tertiary axis is extremely speculative, and will require detailed geomorphological and sedimentological studies for verification. Moreover, the following intriguing relationships should be taken into account:

- (i) The north-easterly portion of the Griqualand-Transvaal axis runs parallel and close to a major anticlinal axis which divides the 2.0 b.y. Transvaal Supergroup into a major northern and a subsidiary southern tectonic basin. The upwarp axis also crosses the Halfway House granite batholith, which was interpreted by Brock (1964) as being a rising continental block during Witwatersrand times (2.3 - 2.4 b.y.).
- (ii) The northeast section of the Escarpment axis runs close to and roughly parallel to the axis of the Lebombo monocline which represents a line of early Jurassic warping (Du Toit, 1930). Along the south coast, this axis runs close to and parallel to the axis of maximum subsidence of the palaeozoic Cape Basin.
- (iii) The Khomas axis recognized by Du Toit (1933) lies along a line of late-Precambrian to early Cambrian uplift (Haughton, 1969).
- (iv) Present-day seismicity in northern Botswana suggests

that the Okavango axis is still active.

Whether these relationships are coincidental, or indicate that epeirogenic uplift took place along similar axes over long periods of geological time, or that the axes are all of different ages, is not yet clear, but obviously fundamental to understanding epeirogenic processes. However, many of the 90 m.y. kimberlites are located in the broad zone between the Griqualand-Transvaal and Escarpment axes (Fig. 6.5). Volcanism may thus have been related to sub-lithospheric stresses associated with an episode of Turonian uplift along these axes, by analogy with the model proposed by Moore (1976). Similarly, early Tertiary and Oligocene volcanics are closely spatially associated with the west coast Escarpment axis, and volcanic activity may have been triggered by uplift along this axis. It is possibly significant that younger volcanic pipes appear to be concentrated close to the continental margin (Davis, 1977).

Factors determining epeirogenic uplift are poorly understood. However, although data are scarce, it would appear that periods of continental uplift may bear a temporal relationship to variations in spreading rates at the northern mid-Atlantic ridge. Periods of slow seafloor spreading are illustrated in Fig. 6.1 (data from Coney, 1971; Pittman and Talwani, 1972; Hays and Pittman, 1973 and Rona, 1973). These show a rough correspondence with episodes of continental uplift and volcanism. If significant, these correlations suggest that epeirogenesis and volcanic activity on the sub-continent are geanti-

cally related to more widespread but poorly understood earth processes.

It is interesting to speculate on the contrasting magnitude of the early Cretaceous alkaline complexes that define prominent NE-trending igneous lineaments, and the younger kimberlite and olivine melilitite pipes. The former are typically large intrusions, several kilometres across (Martin, *et al.*, 1960; Mathias, 1974), and have clearly entailed the emplacement of large volumes of magma. Kimberlites and related pipes on the other hand are far more numerous and more widespread, and individual pipes appear to have been associated with relatively small volumes of magma. This would appear to be true of even very large kimberlites (e.g. the Mwadui pipe in Tanzania, Edwards and Hawkins, 1966) for at depths of roughly 2 km beneath the palaeo-land surface, feeder pipes are typically only tens of metres across (Hawthorne, 1975).

One possible explanation for such contrasting styles of volcanism is that the pattern of stresses which trigger igneous activity in part controls the volume of magma emplaced at any given focus. Thus, where stresses are concentrated along narrow linear zones, these form favourable loci for the accumulation of large magma volumes, and lead to the emplacement of large-scale sub-volcanic complexes. However, where stresses are dispersed as a result of uplift over broad linear zones, magma accumulation is not as efficiently accomplished, and the result is a large number of relatively small volcanic pipes.

In Section 4, it was suggested that in the presence of CO₂, more magnesium-rich magmas are derived by melting at greater depths (and consequently higher temperatures) than Mg-poor liquids. This would account for the progressive increase in MgO content from the Garies volcanics through those in the Gamoep cluster to kimberlites. Reference to Figure 6.4 shows that this sequence of increasing MgO content is from the coast inland. If alkaline magmas are derived from the low velocity zone (Green, 1971) this suggests a thickening of the plate towards the interior of the continent resulting in a central keel. This is consistent with the model proposed by Nixon *et al.* (1973), who believe that sheared nodules in northern Lesotho are derived from greater depths than those in the East-Griqualand area, which is closer to the eastern continental margin.

The discussion in this section has stressed the genetic relationship between alkaline volcanism and epeirogenesis in southern Africa. However, the model involving upwarping of the continent is not proposed as a unique "trigger" to alkaline volcanism, but should be regarded as complementary to the processes suggested by Marsh (1973), and also the "membrane tectonics" model put forward by Turcotte and Oxburgh (1975). These all essentially reflect variations on a tectonic theme involving lithospheric mechanics. Further, the factors determining epeirogenic uplift are poorly understood, and beg refinement to our understanding of plate tectonics processes.

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