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Rb-sr isotopic studies relating to problems of geochronology on the Nelspruit and Mpageni granites

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The theory behind the total rock method is that if radiogenic Sr is lost from some of the minerals, it may remain close to them in the rock trapped in different mineral lattices or in crystal interspaces, and the whole rock satisfies the closed system requirements even though the constituent minerals do not. However, cases have been recorded where even the total rock samples have not remained closed systems (e.g. Lamphere et al, 1963).

The isochron method is applicable, as shown before for the separated minerals, if samples with a sufficiently wide range of Rb/Sr ratios are available, and better precision is achieved if the points are further apart. Samples with a range of Rb/Sr composition can usually be obtained from a suite of rocks of the same age, even though they have been isotopically homogenized.

The total rock method has not completely superseded the separated mineral one. The latter can be used to investigate the metamorphic history after the last complete homogenization of the rock suite. If the rock has not undergone any metamorphic event after homogenization, the mineral ages will be concordant and will lie on the total rock isochron. However, if movement of radiogenic Sr has taken place, the mineral points will lie above or below the total rock isochron depending on whether the mineral phase has gained or lost radiogenic Sr. In this case mineral ages will be discordant if calculated assuming the same initial ratio as the total rock sample. As it is usually impossible to determine how much radiogenic Sr a mineral has gained or lost, mineral ages are then not significant. Biotite, however, is an exception because it readily loses radiogenic Sr^{87} and under special conditions can lose it almost completely (Hart, 1964). Under such conditions biotite ages, calculated assuming as initial ratio the one determined from total rock samples, are indicative of the possible time of metamorphism.

3. CHOICE OF THE METHOD

In the present investigation the Rb - Sr method was chosen for the following reasons.

The rocks studied are very old and from the geological descriptions of the area it appears that a series of granite intrusions and probably different periods of granitization have occurred. In these conditions it can be expected that the rocks have undergone a complex metamorphic history after crystallisation; and that radiogenic nuclides have diffused out of their original crystals because of periods of heating connected with younger intrusive phases.

Rb and Sr, being lithophile elements, readily substitute for K or Ca in the crystalline lattices of the most common rock-forming minerals, and therefore they are less susceptible to diffusion or low temperature hydrothermal migration than U, Pb and Ar over long distances. This suggests that the Rb - Sr whole rock method has the best chance of giving results unaffected by metamorphism. Some information on the metamorphic history of the area can be obtained from the apparent mineral ages but these probably represent maximum ages for the last metamorphic episode.

From Rb - Sr studies it would theoretically be possible to show whether the Mpageni granite is derived from the same source region as the Nelspruit granite if their relative ages, primary Sr^{87}/Sr^{86} ratios and Rb - Sr composition of the two rocks were known accurately. Since observed differences in Sr^{87}/Sr^{86} primary values are usually small, great precision is required in measurements used in this connection.

An additional advantage of the Rb - Sr method is that it can give some information on whether the rocks studied are of crustal or subcrustal origin. From the work of Turekian and Wedepohl (1961) it appears that Rb, K, Na and to some extent Al in the earth's rocks, increase in concentration as the Si content rises. The Sr concentration also increases with the Si but after a maximum in andesites and diorites, drops rapidly in granodiorites and rocks with higher Si content. The U and Pb however, increase with Si both at the same rate. Therefore, while the Rb/Sr ratio

increases from about .1 in basalts to 1.0 and more in granitic rocks, the U/Pb ratio remains constant at about .2 This is an important property because it implies that the more basic rocks of the Upper Mantle have smaller Rb/Sr ratios and therefore the growth of Sr^{87} in respect to Sr^{86} is much slower there than in the granitic crust. This has also been suggested by the analysis of basalts that are thought to have subcrustal origin (Faure & Hurley, 1963; Hedge & Walthall, 1963; Hamilton, 1963; McDougall et al, 1963; Gast et al, 1964; Lessing & Catanzaro, 1964). Heier, Compston and McDougall (1965), have plotted the samples of dolerites more likely to have subcrustal origin in a histogram showing the frequency of occurrence of the initial ratios from several investigations. This histogram shows that most of the dolerites' initial ratios fall between .702 and .706 It seems that the Sr^{87}/Sr^{86} ratio for subcrustal rocks, has remained close to .704 As pointed out by Hurley et al (1963), and Faure & Hurley (1963), the difference in initial Sr^{87}/Sr^{86} ratios between crustal and subcrustal rocks can be used as a criterion to decide on the source of the rocks studied.

Hurley et al (1963), have proposed, on the strength of Rb - Sr data, a model for continent formation by continual addition of mantle material to the crust and have pointed to the low Sr^{87}/Sr^{86} ratios of the rocks studied as proof.

It would be of interest to study all the granites of the Barberton Mountain Land from the point of view of their initial ratios - and therefore their origin, since they are among the most ancient crustal rocks known.

A possible disadvantage of the Rb - Sr method is that the half life of Rb^{87} is not known very precisely. However, the relative age of various formations studied is independent of the half life and the data listed can easily be adjusted if the value of the half life is more accurately known in the future, and if it should differ from the value of $5.0 \pm 0.02 \times 10^{10}$ years used in this investigation.

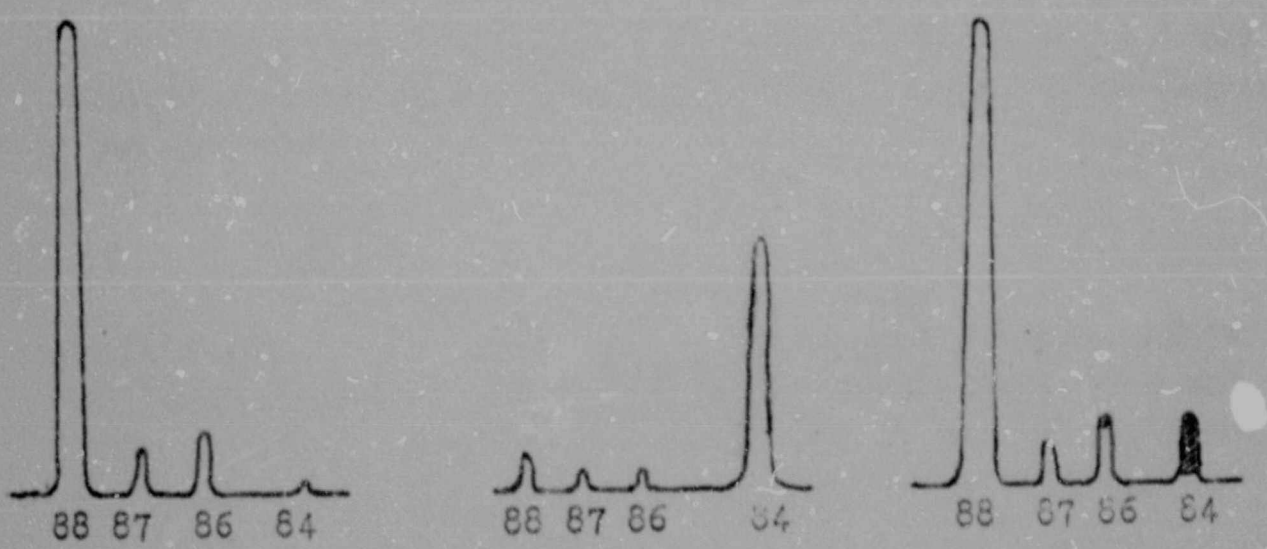
4. THEORY OF ISOTOPE DILUTION TECHNIQUE

Isotope dilution was used to find the concentrations of Rb and Sr isotopes in the rocks from isotopic ratios measured with a mass spectrometer. This was performed by adding to the rock solution accurately known amounts of tracer (or "spike") that consists of a solution of known but unnatural isotopic composition.

In the case of Sr (see fig. 7) the concentration of Sr^{86} was obtained by using a tracer composed mainly of Sr^{84}.

Fig. 7

Strontium Spectra



natural Sr spike Sr spiked sample

In natural Sr the ratio Sr^{84}/Sr^{86} is known to be approximately 1/20. If the ratio is altered by adding to natural Sr a known amount of spike, the concentration Sr^{86} can easily be calculated as follows:

let r = ratio Sr^{84}/Sr^{86} in the rock, constant in natural Sr
s = Sr^{84}/Sr^{86} in spike
m = measured Sr^{84}/Sr^{86} in the mixture
Cr = concentration of Sr^{86} in rock - unknown
Cs = concentration of Sr^{86} in spike - known

Wr = weight of the rock sample
Ws = weight of the spike

then the measured ratio would be:

$$m = \frac{r\ Cr\ Wr}{Cr\ Wr} + \frac{s\ Cs\ Ws}{Cs\ Ws} \qquad (1)$$

and thus:

$$Cr = \frac{Cs\ Ws}{Wr} \cdot \frac{s - m}{m - r} \qquad (2)$$

The quantity $\frac{Cr\ Wr}{Cs\ Ws} = \frac{s - m}{m - r}$ was defined as Q.

Jamieson & Schreiner (1956), have shown that the fractional error in the measurement of Q depends on the value of m and that for some value of m it has a minimum. The "error magnification" is:

$$\frac{\Delta Q/Q}{\Delta f/f} \qquad (3)$$

in the case f = m, equation (3) can be used to investigate the "magnification" for errors in the measurement of s and r. The limit of equation (3) for $\Delta f \longrightarrow 0$ is the multiplicant of any error made in the measurement of m. It will be:

$$Fm = \frac{m(r - s)}{(r - m)(m - s)} \qquad (4)$$

Fm has a minimum at:

$$m^2 = rs \qquad (5)$$

Plots of equation (4) for varying values of m were constructed for Rb and Sr. The curve representing the "error magnification" was in both cases nearly flat for a range of values of m on either side of $m = \sqrt{rs}$ (plate 1). When possible the samples were spiked to obtain values of m close to one. In this way "error magnification"* (see plates 1 & 2) is small, and in addition both instrumental errors (arising from the non-linearity of the amplifier or the recorder) and errors in measuring peak heights on the chart, are minimized.

* in this laboratory

Plate 1 - Sr error magnification plot

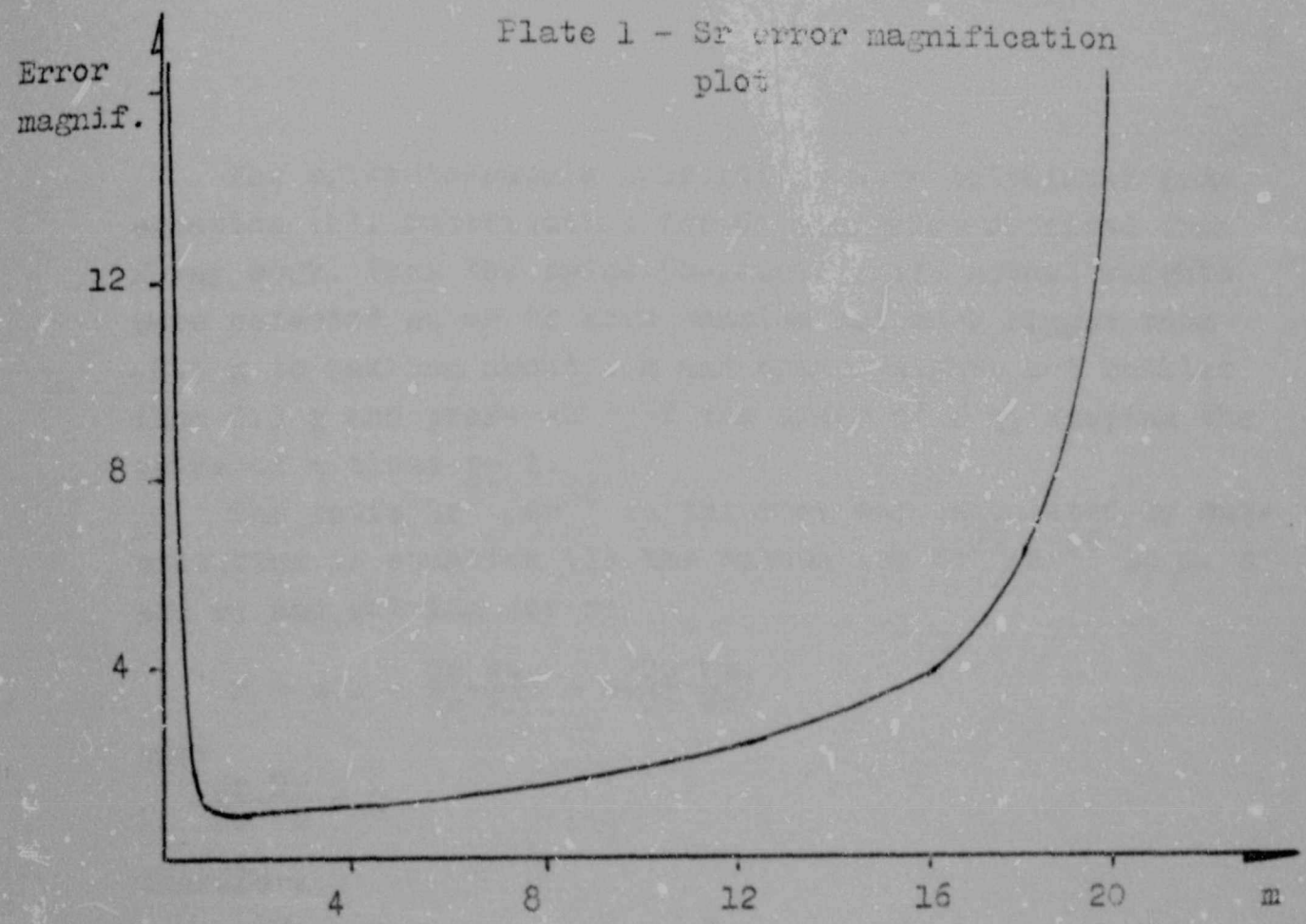
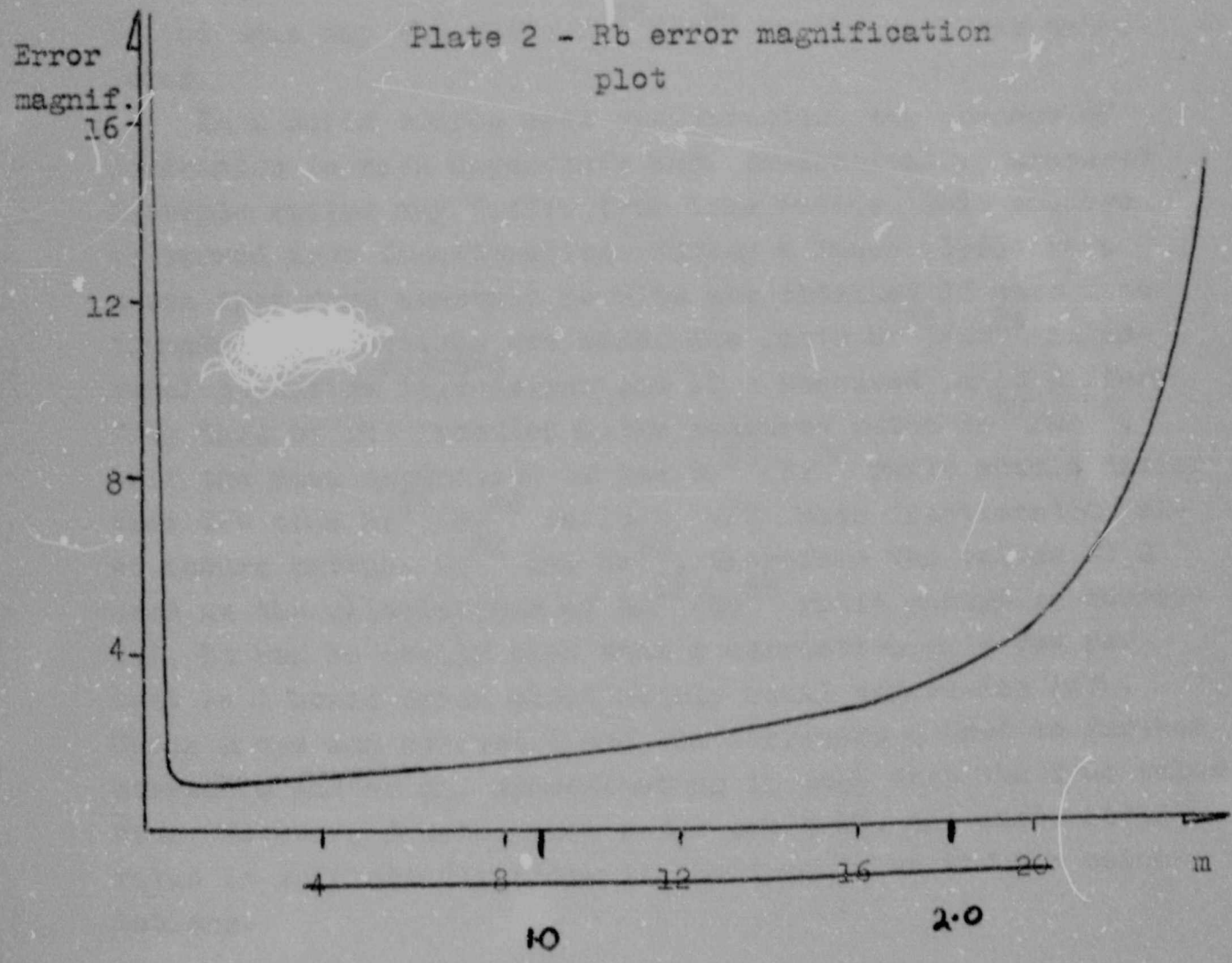


Plate 2 - Rb error magnification plot



The spike-to-sample proportions were calculated from equation (2), substituting for Cr the value obtained from X-ray work. From the spike-to-sample ratio actual weights were selected so as to have samples not much bigger than 0.5 g to maximum about 1 g and spike weights not smaller than 0.3 g and preferably of the order of 1 g, keeping the value of m close to 1.

The ratio Sr^{87}/Sr^{86} in the rock was calculated by substituting in equation (1) the values for Sr^{87}/Sr^{86} in m, s and r; and solving for r:

$$r = m\left(1 + \frac{Cs\ Ws}{Cr\ Wr}\right) - s\left(\frac{Cs\ Ws}{Cr\ Wr}\right)$$

but:

$$\frac{Cr\ Wr}{Cs\ Ws} = Q$$

therefore:

$$Sr^{87}/Sr^{86})_r = m\left(1 + \frac{1}{Q}\right) - \frac{s}{Q}$$

In the same way the ratio Sr^{88}/Sr^{86} in the rock was calculated.

In a solid source mass spectrometer, the process of ionization is mass dependent; and, theoretically, measured isotopic ratios may differ from true ratios. This process is termed mass fractionation. Hurley & Faure (1963) have shown that more constant results are obtained if mass fractionation corrections are made. The ratio Sr^{88}/Sr^{86} in natural strontium is assumed constant and if a measured ratio differs from this by the fraction Δ, the measured ratio Sr^{87}/Sr^{86}, half the mass separation of the Sr^{88}/Sr^{86} ratio should differ from the true Sr^{87}/Sr^{86} ratio by $\Delta/2$. Mass fractionation also occurs between Sr^{84} and Sr^{86}, therefore the values of Q used in the calculations of Sr^{88}/Sr^{86} ratio should be corrected. It can be easily seen that a correction of a few per cent in Q would bring about nearly equal correction in Δ. Using Δ one can correct Q and the corrected Q used to further correct Δ and so on, approximating in each step the true value of Δ. However, Δ being only a few per cent, the uncorrected value is sufficiently close to the true value for our calculations.

The value $Sr^{86}Cr$ is also affected by mass fractionation but no correction was applied because it was not possible to measure the Δ in the case of Rb, and it was hoped that this value would be comparable with that of the Sr and that the omission of both fractionation corrections would not affect the Rb^{87}/Sr^{86} value.

From fifty measurements on Sr samples it was found that the average fractionation effect between Sr^{88} and Sr^{86} was of the order of 1.4%. Only in four cases was it as high as four per cent or more, with a maximum value of 4.8 per cent. In a few cases the concentration of the isotope of heavier mass decreased during the run in respect to the lighter isotope. In these case the fractionation was called negative and correction was applied in the opposite direction.

CHAPTER II - THE GEOLOGICAL SETTING

1. INTRODUCTION

The Barberton Mountain Land is composed of one of the oldest suites of sedimentary and volcanic rocks surrounded by various bodies of granitic rocks.

Attention was drawn to the geology of this area by the discovery of gold mineralization in the second half of the 19th Century. A full account of the structures and stratigraphy was published by Hall in 1918.

Following Hall's work very little was published on the geology of this area until the South African Geological Survey started new investigations in 1937. The Survey's volume on the Barberton Mountain Land was published in 1956.

Since then a new interest in the very complex geology of the area has grown among South African scientists and several scientists from other countries, especially the United States and Britain, have had their attention drawn to the Barberton Mountain Land because of its interesting geology. These rocks, being so old, yield valuable information as regards the early stages of evolution of the earth's crust and the physical conditions of the planet at approximately one quarter of its present age.

2. SIMPLIFIED DESCRIPTION OF THE GEOLOGY OF THE MOUNTAIN LAND

The stratified rocks of the Swaziland System:

The old sedimentary rocks are grouped under the name of Swaziland System. This has been sub-divided in three series so that the stratigraphic column is:

SWAZILAND SYSTEM	MOODIES SERIES	- conglomerates, sandstones, shales
	unconformity	
	FIG TREE SERIES	- shales and graywackes
	ONVERWACHT SERIES	- acid lavas, basic lavas
BASEMENT		- gneisses ? granites ?

Basement: The basement on which the Swaziland rocks have been deposited is not exposed. It is, in general, believed to be of granitic nature. Anhaeusser & Viljoen (1965) are of the opinion that the typical Nelspruit migmatite repre-ents, for the most part, the basement upon which all the layered rocks of the Mountain Land were deposited. Van Eeden & Marshall (1965) however, are against this hypothesis and propose instead that the Nelspruit granite derives from granitization of rocks of the Swaziland System up to the Moodies series, possibly down to sediments older than the Onverwacht, and possibly even a granitic basement rock.

Onverwacht Series: Visser et al (1956) describe the Onverwacht rocks as a succession of basic lavas overlaid by a succession of acid extrusives. The basic lavas are of andesitic composition, fine grained in texture, but they can vary to a dark green aphanitic variety. Flow layers can still be recognised in the less altered lavas. Amygdaloidal varieties are also present, and in some hand specimens tiny gas vesicles filled with pink calcite can be observed. The acid lavas are of rhyolitic composition and can be divided into porphyritic varieties and dark, dense felsite.

Gribnitz et al (1961) have found dolomitic and arenaceous rocks, banded cherts and banded ironstones which are

pre-Fig Tree in age, and they have proposed the existence of an older sedimentary formation which they have named Oorschot. They believe that the rocks of the Onverwacht are intrusive in the Oorschot series.

Anhaeusser & Viljoen (1965) have proposed that these rocks of the Oorschot should be included in the Onverwacht together with a good portion of the metamorphic rocks of the aureole of the Nelspruit Granite which are classified as Jamestown.

The inclusion in the Onverwacht of the sediments of the Oorschot series and a large amount of the igneous rocks of the Jamestown , makes this assemblage virtually identical to that belonging to the initial magmatic phase of a typical geosyncline (Roering, 1965).

Fig Tree Series: The Fig Tree series is composed mostly of argillaceous sediments, but coarse, clastic sediments - notably bands of quartzite, grit conglomerate and beds of graywackes - also occur. At the top of the sequence there are rocks that have been interpreted as acid lavas (Visser et al, 1956), or as rocks formed by tuffaceous and lava materials (Anhaeusser & Viljoen, 1965), or as graywacke slump breccia (Ramsay, 1963).

Shales, banded cherts and banded ironstones are the major rock constituents in the Fig Tree series and they are so closely banded that Visser et al (1956) did not make quantitative distinction between them in the field.

Roering (1965) has interpreted the sedimentary structures as indicating that the graywackes have been deposited in deep water by turbidity currents and are therefore similar to flysch sediments, while the presence of chemical precipitates like cherts, jaspers and ironstones represents a period of calm sedimentation.

Moodies Series: The Moodies rocks rest uncomformably on older rocks, and were considered by Visser et al (1956) as a separate system from the Swaziland rocks. By the recovery of deformed Fig Tree pebbles in the Moodies conglomerates, they

also suggested a period of deformation younger than the deposition of the Moodies rocks.

The Moodies, unlike the Fig Tree, are shallow water sediments and probably mark the late stage of the Swaziland geosynclinal deposition and for this reason they have been included in the Swaziland System. The Moodies series is composed of coarse sediments: boulder beds, conglomerates, sandstones and quartzites, alternating with beds and lenses of fine and very fine material: siltstones, shales and locally, ironstones and jaspers. Cross bedding and ripple marks are common features in this type of sediment and mud cracks are reported to occur in the fine grained rocks (Visser et al, 1956; Ramsay, 1963). The base of the Moodies is marked by a conglomerate which contains some granitic boulders and pebbles of unknown origin.

The Intrusive Rocks:

The stratified rocks of the Swaziland System are surrounded on all sides by granitic rocks. A feature common to all the granitic rocks is their intrusive contact with the Swaziland System. Other intrusive rocks comprise the ultrabasic assemblage of the Jamestown complex and a multitude of basic dykes ranging in age from prior to the granitization to Karoo age.

The Swaziland granites situated on the south-eastern side of the Barberton Mountain Land, have been studied extensively by Hunter (1957, 1961, 1965), who has introduced three main divisions in the granitic rocks in relationship to the orogeny of the Swaziland System:

- the pre-orogenic gneisses (basement?)
- three synorogenic cycles of granite emplacement
- two postorogenic cycles.

A number of papers have been written on the granites in the north, south and west of the Barberton Mountain Land the more recent of which are: Visser et al (1956), Ramsay (1963), Viljoen (1964), Anhaeusser (1964, 1966), Anhaeusser & Viljoen (1965), Van Eeden & Marshall (1965), Engel (1966), Roering (1967).

All these granitic rocks are believed to have intruded

in post Moodies time. The Nelspruit granite and the Kaap Valley granite (see map 2) have acted as controls of the deformation and structures can be traced along the contacts. These plutons can probably be classified as synorogenic although some authors (Anhaeusser & Viljoen, 1965) tentatively recognize the Nelspruit granite as the basement of the stratified series.

Younger intrusives like the Mpageni granite, the Dalmein and the Salisbury Kop granite are discordant to the structural trends and can be classified as postorogenic.

Roering (1967) has advanced the hypothesis that the granitic rocks have non-orogenic origin. They have been the cause of the folding but are not intimately associated with it. This implies an independent process of granitic formation.

Engel (1966) has pointed to continuous degassing of the mantle as possible origin of the large quantities of granitic rocks. This mechanism of formation could be checked by isotopic studies and will be discussed at a later stage.

The Nelspruit Granite:

According to Van Eeden & Marshall (1965) "Nelspruit Granite" is a "sack" name given to an assemblage of rocks derived from the granitization of a large number of different rocks ranging from the Moodies Series to probably pre-Onverwacht rocks. The Nelspruit granite therefore includes a number of phases ranging from "partly granitized rocks to mobilized granitic material intrusive in ungranitized rocks", and it has variable chemical composition. For this reason, only the portion of Nelspruit granite included in the area covered by the present investigation will be described in some detail.

The area studied extends from the contact between the Nelspruit granite and the metamorphic rocks of the Swaziland System immediately north of the Consort Mine to the northeast, including the Mpageni granite, and to the west, including the Lowveld Crusher's Quarry situated about two miles west of Nelspruit (see map 3). This area has not been mapped

by the author but some traverses were performed for sample collecting and it has been described in part by Visser et al, (1956), Anhaeusser (1964) and Viljoen (1964) in their unpublished M.Sc. theses, and in Information Circulars of the Economic Geology Research Unit nos. 25 (1965) and 32 (1966). From field observations and from the quoted descriptions it appears that the portion of Nelspruit granite studied is a migmatite as defined at the Symposium of Migmatite Nomenclature: (a) "composite, heterogeneous rocks, consisting of pre-existing rock material and of granitic material, subsequently intruded or originated in situ." (Polankov)

(b) "megascopically composite rocks, that once consisted of geochemically mobile and immobile (or less mobile) parts" - (Dietrich, Menhert)

(International Geological Congress, Copenhagen, 1960). Menhert (cf. op.cit.) designates as mobilization the increase of geochemical mobility of rocks or portion of rocks beyond the dimensions of the individual crystals, regardless of the state of aggregation of the rocks and the mechanism of migration. This part of the Nelspruit granite will therefore be referred to as the "Nelspruit migmatite".

The Nelspruit Migmatite:

The Nelspruit migmatite is a composite rock consisting of alternate bands of more felsic and more mafic composition. The more felsic bands are granitic in composition and vary considerably in texture from place to place being granitic, porphyritic or aplitic. These represent the "granitic material" of Polankov (cf.op.cit.) or the "mobile part" of Dietrich and Menhert (cf.op.cit). A common feature is the biotite enrichment at the border of the felsic bands. This could point to a replacement origin of the felsic minerals and has been interpreted as a "basic behind" (Read, 1956 p.353) or restit. The origin of granitic material however, is a very controversial point and is therefore left for more specialized studies.

The more mafic phases contain more biotite iron minerals and in places (close to contact with metasediments) hornblende.

They represent the less mobile phase and probably derive from pre-existing rocks.

The principal mineral constituents of the rock phases described are oligoclase, K-felspar and quartz, together with biotite-magnetite and accessory zircon, sphene and apatite etc. The percentage of the major constituents varies between the different bands or flow layers. Lenses and bands vary in thickness from a fraction of an inch to a few feet; thin felsic bands are very numerous in places, giving the rock a characteristic striped appearance (photographs 1-4). In places, however, the felsic components are predominant and the rock has the appearance of homogeneous granite, for example, an elongated irregular body of "homogenous granite" more than a hundred feet wide which was sampled about half a mile north of the main granite contact, north of the Consort Mine - sample AA7.

The grain size is very variable. In some places the rock is fine grained and - more frequently - medium grained; in other places it becomes coarse grained. The migmatite has undergone deformation and the "flow layers" are in places contorted. The folding, as to be expected in this type of rock (King 1963) is generally irregular and complicated but Visser et al (1956) have found that away from the contact the "flow layers" trend at 55° - 60° east of north, roughly parallel to the strike of the major fold axis of the Mountain Land.

Pegmatites are frequent and occur both parallel and at an angle to the layering. The pegmatites encountered in the present investigation can be divided roughly into two groups: the ones further away from the contact are composed mostly of microcline and quartz with small amounts of biotite; closer to the contact and intrusive in the metamorphic rocks, adjacent to the migmatite, occur pegmatites that are rich in muscovite and show a higher Rb/Sr ratio.

The origin of the Nelspruit migmatite has been discussed in several papers besides the one already mentioned by Van Eeden & Marshall (1965). Read (1956) proposed that the migmatite was caused by granitization of pelitic and more siliceous rocks. The ultrabasic rocks of the Jamestown schist

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