THE ENDOGENOUS CONTACT-ZONE OF THE MAGNESIAN LIMESTONES AT BEN BULLEN, N.S.W.

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(With five text-figures.)

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INTRODUCTION.

Ben Bullen is situated on the Mudgee-Wallerawang railway line at a distance of 121 miles from Sydney.

The general geology of the area has been described by Carne⁽¹⁾ and Carne and Jones,⁽²⁾ and they have shown that a small boss has invaded a sedimentary series consisting of quartzites, tuffs, shales and limestones. The plutonic complex shows a range from quartz-mica-diorite to olivine-norite, and the limestones contain magnesian and non-magnesian types.

The writer⁽⁸⁾ has recently described contaminated rocks which have been formed by reaction between the nonmagnesian limestones and the intrusive diorite. In the present paper the types produced by the assimilation of magnesian limestones are described.

FIELD RELATIONS.

Magnesian limestones occur in the Main Quarry in Portions 45, 46 and 47, Parish of Ben Bullen. Nonmagnesian types are often interbedded. The limestone mass is somewhat lenticular in shape and the longer axis measures $4\frac{1}{4}$ chains. On the north, south and west it is surrounded by quartz-mica-diorite, and on the east alluvium and quarry *débris* obscure all field relations.

A wide zone of contaminated rocks surrounds the limestone mass (see Fig. 1), and gradually merges into the diorite. Quarrying has extended into the inner margin of this zone, and the section (Fig. 2) shows that the field



Fig. 1.

relations are very complicated within this area. Numerous sills and tongues of contaminated igneous rock pierce the limestone, and it is impossible to measure distances from

any one contact. It is obvious that the contaminated zone may be divided roughly into an inner and an outer ring. The inner part is essentially a metasomatized limestone, and in this serpentine-, chlorite-, prehnite- and actinolite-bearing types occur as veins and irregular masses. The outer zone has the appearance of a light-coloured, coarse-grained diorite, and is characteristically a contaminated igneous rock. Veins of calcite and serpentine often occur in the outer zone.

The boundaries of the limestone mass are more complicated than shown in the map (Fig. 1), but in a general way this indicates the disposition of what can be recognised in the field as a contaminated igneous rock and a metasomatized limestone.



Fig. 2.

Section of the face of the Main Quarry. This was drawn about twenty years ago when quarrying was in operation. (From *Geol. Surv. N.S.W.*, Min. Res. 25, p. 316.)

To the north of the quarry the limestones are less magnesian, and on the northern slope of the hill veins of andradite skarn and nodular masses of hæmatite have been noted.

PETROGRAPHY.

(i) The Magnesian Limestones.

Calcite-dolomite-marbles, brucite-marbles and forsteritemarbles occur in the Main Quarry, and in these the magnesia content is very variable.⁽⁹⁾ No true dolomites are known, but it is possible that they occur in restricted bands.

Analyses of the magnesian limestones from this locality are given below.

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	I.	II.	III.
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CaCO ₃	96.76	95.60	85.85
MgCO ₃	$2 \cdot 46$	$3 \cdot 42$	12.10
MnCO ₂		0.06	0.07
$Fe_2O_3 + Al_2O_3 \dots$	0.22	0.18	0.58
P.O		0.03	Judia - Tool
Gangue	0.50	0.96	1.34
	99.94	100.25	99.94

I. Forsterite-spinel Marble, Pors. 45, 46 and 47, Ph. Ben Bullen, Geol. Surv. N.S.W., Min. Res. 25, p. 378.

II. Forsterite Marble, Top Quarry, Por. 47. *Ibid.* III. Sampled Specimen (mixture of Brucite-forsterite-spinel Marble and Calcite-dolomite Marble), Main Quarry, Por. 45-46. Ibid.

(ii) The Quartz-Mica-Diorite.

This will be dealt with more fully when the plutonic complex is described in a later publication. The rock is hypidiomorphic granular, subophitic to poikilitic, with a grainsize averaging 2 mm. The constituent minerals are plagioclase, hornblende, biotite, quartz, iron ores and apatite. Small quantities of epidote, prehnite, chlorite and white mica may occur as alteration-products. The plagioclase varies from $Ab_{52}An_{48}$ to $Ab_{59}An_{41}$, and occasional zoned phenocrysts show a range from Ab49An51 to Ab₆₃An₃₇. Green hornblende has a mean refractive index of about 1.670. The chemical composition is :

SiO ₂				$55 \cdot 30$
Al ₂ O ₃				$20 \cdot 87$
Fe ₂ O ₃				$3 \cdot 62$
FeO				$5 \cdot 20$
MgO				2.75
CaO				7.44
Na _o O				$2 \cdot 49$
K _o Õ				0.82
$H_{0}O +$				0.76
H.O				0.08
TiO				0.90
P.O.				0.41
MnŐ				0.10
				100.74
Sp. Gr.	1.0	and the	1410	2.83

Sp. Gr. Anal. G. A. Joplin.

1000 - 100 - 100 - 100 - 100 - 100 - 100 - 100 - 100 - 100 - 100 - 100 - 100 - 100 - 100 - 100 - 100 - 100 - 100	Norm.	
Quartz		 16.44
Orthoclase		 $5 \cdot 00$
Albite		 20.96
Anorthite	• • •	 $34 \cdot 19$
Corundum		 $3 \cdot 37$
Hypersthene		 12.05
Magnetite		 $5 \cdot 34$
Ilmenite		 1.67
Apatite		 1.01

(iii) The Contaminated Rocks.

It has already been shown that the field evidence gives no clue as to the degree of contamination, and that the relations between some of the types are unknown. The petrography, however, provides some evidence, and in the following section the various assemblages are described in the order which is believed to represent a gradually increasing lime-magnesia content.

(1) Diopside-bearing Quartz-aiorite and Included Xenoliths.

Rocks of this type occur above the N.W. end of the Main Quarry in Portion 47. The xenoliths vary from $\frac{1}{2}$ " to 6" in diameter and have sharply defined outlines. They are fine-grained, greyish-green rocks with a high density. The rock-mass in which they are included usually has the appearance of a normal diorite, but may be coarser grained, with large hornblende individuals in a ground mass of felspar, quartz and sage-green diopside.

The xenoliths are granoblastic rocks consisting either of a mosaic of diopside and plagioclase or of diopside alone. Occasional patches of quartz occur, and around these the diopside forms slightly larger subidioblastic prisms. "Pools" and veins of plagioclase grains and large poikilitic crystals of hornblende are not infrequent, and their occurrence suggests mechanical introduction from the magma (Fig. 3). Apatite and sphene are usually abundant, and a little iron ore may occur.

The diopside grains average 0.2 mm. The mineral is very pale green, with $\alpha'=1.692$, $\gamma'=1.717$ and $Z \wedge c=43^{\circ}$. This indicates $\text{Di}_{64}\text{He}_{36}$. Plagioclase occurs in small laths or in minute xenoblasts which form a granular mosaic. Twinning is not well developed. The extinction on 010 is $34\frac{1}{2}^{\circ}$, $\alpha'=1.558$, $\gamma'=1.568$, and the composition would thus appear to be $Ab_{40}An_{60}$.

The smaller xenoliths show a great deal of breaking up and veining by quartz and felspar. Resorption is evidently effected by the mechanical invasion of a quartz-plagioclase magma (see p. 150).

The diopside-bearing quartz-diorite, which surrounds the xenoliths, consists of plagioclase, hornblende, diopside, quartz, apatite, sphene and iron ores.

The plagioclase forms tabular crystals which may measure up to 1.5 mm. It is often zoned, and oscillatory zoning is sometimes developed. The bulk of the felspar has an extinction angle of 26° on 010, with $\alpha'=1.552$, $\gamma'=1.560$.

The composition is, therefore, Ab₅₃An₄₇. Plagioclase may show alteration to epidote or clino-zoisite, and small veins of white mica sometimes occur.

The hornblende is rather poikilitic and may include plagioclase, iron ores, apatite and diopside. X=light yellowish green, Y=olive green, Z=olive green; $Z \wedge c=14^{\circ}$; $\alpha'=1.660$, $\gamma'=1.683$.



Fig. 3.

A. Diopside-plagioclase xenolith enclosed in diopside-bearing quartz-diorite. \times 12.

Note diorite at top left with larger crystals of diopside bordering the margin of the xenolith. A poikilitic crystal of hornblende is shown at the bottom of the figure, and just above it an irregular "pool" of plagioclase is fringed with larger crystals of diopside.

B. Diopside-bearing quartz-diorite. \times 12.

Note diopside crystals fringed with amphibole and also independent crystals of hornblende. At the top of the figure amphibole and pyroxene are in parallel intergrowth. Note also large crystal of plagioclase with the centre entirely altered to clinozoisite.

The amount of diopside present is very variable. In one slice it is very abundant and in another, at the same distance from a xenolith, it is but poorly developed. It may form the core of a hornblende crystal, it may show parallel intergrowth with hornblende, or it may arise as

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independent prisms measuring about 0.8 mm. It is pale green, with $Z \wedge c = 44^{\circ}$.

As in the parent quartz-mica-diorite, quartz is interstitial.

(2) Diopside-zoisite-plagioclase Rocks with White Mica.

These rocks occur above the quarry at a short distance south of the diopside-bearing diorite. They are also met with amongst the quarry *débris*.

In the hand specimen they resemble igneous rocks, but are lighter coloured and slightly coarser grained than the normal diorite. White mica may usually be detected in the hand specimen.

Under the microscope they are seen to consist of diopside, β -zoisite, plagioclase, white mica, albite, sphene and a little apatite and actinolite. Small quantities of calcite, chlorite and serpentine are often present in veins.

Diopside forms subidiomorphic prisms measuring about 1.5 mm. and is often twinned. $Z \wedge c = 43^{\circ}$, $\alpha' = 1.688$, $\gamma' = 1.710$. This indicates $\text{Di}_{58}\text{He}_{42}$. Actinolite and calcite are often developed along cracks in the pyroxene.

The labradorite $(Ab_{41}An_{59})$ forms large tabular crystals which measure about 2 mm. In the less altered types it shows flecking with white mica, and a little granular zoisite and albite is often associated. In the more altered types the felspar is entirely replaced by white mica and the felspar areas are now occupied with criss-cross flakes which measure about 0.2 mm. The mica is biaxial and negative, with a fairly small 2V. The elongation along the cleavage is positive; $\alpha'=1.562$, $\gamma'=1.600$. According to Kunitz⁽¹⁰⁾ this indicates an aluminous mica with about 2.5% of iron. The analysis given below is of a completely altered rock very rich in white mica, and the percentage of alkalis would indicate that both muscovite and paragonite are present.

The zoisite forms short subidiomorphic prisms which are aggregated into clusters. These often fringe the diopside crystals (Fig. 4A), but they may occur as isolated masses within the areas of white mica. The zoisite is biaxial and positive, the elongation is negative and the extinction is straight. The birefringence is very low, but the mineral does not show anomalous interference tints. It is therefore β -zoisite which, according to Winchell⁽¹⁴⁾ may contain up to 5% of iron. A little clinozoisite is sometimes associated. Sphene occurs in fairly large idiomorphic crystals or as minute granules; it is usually well developed. Actinolite is present in small amount and may fringe the diopside crystals or occur as small independent prisms. It is usually crowded with granular sphene. Sometimes it occurs in veins. Apatite occurs as small idiomorphic inclusions but is not abundant.

An analysis of the most completely altered rock of this type is given below.

SiO ₂				$47 \cdot 62$
Al ₂ Õ ₃				$18 \cdot 21$
Fe ₂ O ₃				0.72
FeÕ				6.46
MgO				5.47
CaO		N. 19		13.67
Na _o O		61. A 44		2.49
K ₀ Õ				1.16
$H_{0} +$				1.81
$H_0 -$				0.12
TiO.				1.36
P.O.		1-015-01		0.23
MnÖ				0.12
CO.				0.43
Cl	100 100			0.03
	100	1000	1.1	
				99.90
Less O f	or Cl			0.007
1055 0 1	01 01			0 001
				99.89
Sp Gr				3.00
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(3) Actinolite-albite-clinozoisite Rocks.

These rocks occur sporadically at various places within the endogenous contact-zone. In the hand specimen they have the appearance of light coloured rather felspathic diorites. Carbonates are usually present in thin sheets along joint-planes.

Under the microscope the rock is hypidiomorphic granular and the grainsize averages about 2 mm. The constituent minerals are albite, diopside, actinolite, clinozoisite, calcite, sphene, apatite and traces of iron ore. Calcite, acicular actinolite and prehnite occur in veins, and the latter often replaces the albite in the vicinity of the vein. Small patches of spherulitic chlorite may also occur.

Albite forms irregular grains measuring from 0.8 to 3 mm. It is optically positive, with $\alpha'=1.530$, $\gamma'=1.540$, and the extinction on 010 is -13° . The composition is

therefore $Ab_{97}An_3$. The mineral is rather brown and turbid, and encloses granular masses of clinozoisite and occasionally a little epidote. When adjacent to veins of prehnite it is often replaced by clear subidiomorphic crystals of that mineral. Alteration to chlorite is sometimes noted.

Diopside forms subidiomorphic prisms averaging $2 \cdot 5$ mm., and these are often fringed with actinolite. The amphibole may entirely replace the pyroxene and also may arise as independent crystals in the contaminated rock. When primary it occurs in slender prisms measuring about 2 mm. It is optically negative, $Z \wedge c=19^{\circ}$, $\alpha'=1.628$, $\gamma'=1.655$. The colour shows a peculiar patchy distribution and may vary from colourless to green in a single crystal. X=pale yellowish green, Y=pale yellowish green, Z=pale bluish green (Z > Y > X). This corresponds fairly closely to an actinolite described by Ford⁽⁵⁾ which has a mean R.I. of 1.637 and contains about 2.7% of alumina. When the actinolite occurs in veins it is developed in acicular crystals.

Clinozoisite occurs in small granular masses within the albite, but is not abundant. Sphene forms subidiomorphic crystals up to 0.8 mm., and often contains a core of iron ore. In the larger crystals a good cleavage on 110 may be developed.

(4) Actinolite-chlorite-epidote Rocks.

These rocks are essentially similar to those described above, but diopside is absent, actinolite is well developed, epidote is far more abundant than clinozoisite and the place of albite is taken by spherulitic chlorite.

It has already been stated that albite shows incipient alteration to chlorite, and it would appear that this mineral has arisen directly from the felspar. The chlorite has a very low birefringence and shows ultra-blues. It is optically negative and the elongation of the small spherulitically arranged needles is positive. It therefore appears to be pennine.

The actinolite occurs in subidiomorphic prisms from 1 to 3 mm. It is optically negative and the elongation is positive. $\alpha'=1.622$, $\gamma'=1.647$; $\triangle=0.025$; X=very pale yellowish green, Y=pale yellowish green, Z=bluish green (Z>Y>X); Z $\land c=16^{\circ}$. According to Ford⁽⁵⁾ this would indicate a small percentage of alumina. The epidote is well developed in subidiomorphic crystals and a fairly well-marked pleochroism indicates a moderately high iron content. Epidote and actinolite are sometimes intergrown in a mosaic and form small skarn-like patches. A little calcite and sphene may be associated with these patches.

Sphene, apatite and iron ores are accessory, but the first is often well developed.

(5) Actinolite-serpentine-epidote Rocks.

This type appears as veins and irregular dark, finegrained patches in the actinolite-albite-clinozoisite rocks.



D.

Fig. 4.

A. Diopside-plagioclase-zoisite rock with plagioclase entirely replaced by white mica. \times 9.

Zoisite builds prismatic crystals about diopside and also forms larger isolated masses. White mica occurs in criss-cross flakes and sphene surrounds iron ores. Inclusions of apatite occur in the diopside and zoisite.

B. The lower half of the figure represents an actinolite-albite-clinozoisite rock showing turbid albite and small granular clusters of clinozoisite.

The upper part of the figure shows the adjacent actinoliteserpentine-clinozoisite rock occurring as a wide vein in the albitebearing assemblage. Clear plates of serpentine enclose granules of clinozoisite.

Ragged prisms of actinolite and crystals of sphene occur in both rocks. \times 9.

C. Diopside-chlorite-spinel rock. \times 9.

Note large area of chlorite with some sections showing one cleavage. The remainder of the rock consists of a mosaic of diopside and spinel with some apatite. The vein consists of clinozoisite.

It is obvious that the serpentine is taking the place of albite, and at the junction of the two assemblages plates of clear serpentine enclose small, turbid remnants of the felspar. The serpentine has a very low birefringence, it is optically positive, and $\alpha'=1.562$, $\gamma'=1.567$.

Actinolite occurs as in the other assemblages and may show polysynthetic twinning. $\alpha'=1.628$, $\gamma'=1.650$; $Z \wedge c=19^{\circ}$; X=yellowish green, Y=yellowish green, Z=bluish green (Z > Y > X).

Epidote forms granular masses and stout subidiomorphic prisms. It is yellowish green and markedly pleochroic, $\alpha'=1.732$. According to Winchell⁽¹⁴⁾ this indicates $Fe_2O_3=25\%$. Clinozoisite is sometimes associated but is not abundant. Near the junction of the albite assemblage, however, it occurs in small granules and epidote is rare (Fig. 4B).

(6) Prehnite-bearing Rocks.

This type occurs in a wide dyke-like mass associated with serpentine. It is also met with in smaller veins. In the hand specimen it is a streaky pink-and-green rock, and white mica is clearly visible.

Under the microscope the rock shows considerable variation in texture and in the relative proportions of the minerals present. The constituent minerals are prehnite, diopside, serpentine, white mica, sphene, zoisite and sometimes a little albite, chlorite and iron ore. Garnet has been noted in one slice. The prehnite usually forms fairly large interlocking grains, which are brown and turbid and sometimes appear rather fibrous. When the prehnite surrounds areas of serpentine it often develops clear subidioblastic crystals.

The other minerals show characters similar to those in the assemblages described above.

(7) Pyroxene-chlorite-spinel Rock.

Only one example of this type is recorded. It occurs near the edge of the quarry above its north-west end (Fig. 1). In the hand specimen it might be mistaken for an igneous rock, but closer examination shows an abundance of spinel and small areas of calcite. The density is far above that of a normal igneous rock.

Under the microscope the rock shows considerable variation in the distribution of the constituent minerals,

and a mineral which may be very abundant in one slice may be scarcely represented in another cut from the same hand specimen.

The structure is granoblastic and intersertal and the constituent minerals are pyroxene, spinel, chlorite, calcite and apatite. Small grains of iron ore sometimes occur, and a little clinozoisite may be present.

Pyroxene occurs in small xenoblasts, but when it surrounds areas of chlorite or calcite it may develop large subidioblastic prisms. It is very pale green, $Z \wedge c=40^{\circ}$, $\alpha'=1.700$, $\gamma'=1.720$. Its association would suggest that it contained alumina.

Spinel is well distributed throughout the rock, but may show local segregations. It often occurs in large arc-shaped aggregates of small grains, or several large crystals may form a compact mass measuring about 3 mm. across. The usual occurrence is as small xenoblasts in a mosaic of pyroxene. When adjacent to areas of chlorite or calcite subidioblastic outlines may be developed and the spinel often shows a narrow rim of serpentine which separates it from calcite. The spinel is bright green, but the depth of colour is variable; n=1.750. The spinel sometimes shows a separation of finely-divided iron ores. Larsen and Berman⁽¹¹⁾ quote a green spinel with a R.I. of 1.730; this contains ferric iron and some alkalis.

Two varieties of chlorite are developed, but one is much more abundant than the other. It is usually interstitial, and single crystals may measure up to 4 mm. One good cleavage is developed (Fig. 4c) and the extinction is parallel to this. The well-developed chlorite is optically negative, with a fairly small 2V. The elongation is positive; X =pale brown, Y = pale green, Z = pale green (X > Y = Z); $\alpha' = 1.630$, $\gamma' = 1.645$. According to Winchell⁽¹⁴⁾ this indicates a ferroantigorite with about 20% daphnite and 20% of antigorite.

The less well-developed chlorite is optically positive, the elongation is negative and the R.I. is lower than the above. It is evident that it contains more alumina and less iron and magnesia.

Calcite is developed in large irregular grains and is usually interstitial. In some of the slides it is not represented, and in others it is very abundant. It may occur in small grains in a granular mosaic with diopside. Apatite is also sporadic in its distribution. When present it usually forms subidioblastic crystals up to 0.5 mm. These have a

somewhat stout prismatic habit and may form small clusters of two or three crystals.

An analysis of this rock is given below.

SiO ₂		 	$29 \cdot 66$
$Al_2 \tilde{O}_3$		 	$24 \cdot 51$
Fe ₂ O ₃		 	$3 \cdot 99$
FeO		 	$3 \cdot 27$
MgO		 	$14 \cdot 56$
CaO	•	 	18.05
Na ₂ O		 	0.14
K2Ō		 	0.13
$H_{0} + 0$		 	$2 \cdot 02$
$H_{2}O -$		 	0.05
TiO ₂		 	0.90
P_2O_5		 	0.31
MnŐ		 	0.17
CO ₂		 	$1 \cdot 91$
201			

99.67

Sp. Gr. .. $3 \cdot 29$. . Anal. G. A. Joplin.



Fig. 5.

A. Actinolite Skarn. \times 12.

Note radiating needles of actinolite which pierce large irregular grains of calcite, and vein-like occurrence of iron ore.

B. "Pegmatite." \times 12.

Large allotriomorphic grains of quartz and calcite with inclusions of actinolite, diopside, sphene and iron ores. Small masses of chlorite are interstitial.

(8) Actinolite Skarn and Associated "Pegmatite".

This type occurs both in the Main Quarry and in the quarry on Cleared Hill. The "pegmatite" is ore-bearing and occurs as a coarse-grained dyke rock. The skarn forms a border about three inches wide on either side of the dyke, and crystals of actinolite penetrate the adjacent limestone.

The skarn consists of actinolite, quartz, calcite, sphene, iron ore and a trace of epidote. The actinolite is very abundant and forms large fan-shaped masses of radiating crystals which may measure up to 1'' in length (Fig. 5A). Calcite and quartz are developed in large (6 mm.) grains, and no reaction is apparent at the junction of the two minerals.

Both calcite and quartz enclose needles and radiating mats of actinolite. Granular sphene, epidote and iron ores are also included.

Actinolite is optically negative; the elongation is positive; X=colourless, Y=Z=yellowish green; $\alpha'=1.631$, $\gamma'=1.660$. According to Ford⁽⁵⁾ this indicates about 4.4% of alumina. Actinolite and iron ores often form intergrowths from which quartz and calcite are absent.

Iron ores consist of both magnetite and pyrites, and the former shows alteration to hæmatite.

The "pegmatite" consists of large (6 mm.) grains of calcite and quartz (Fig. 5B). These contain inclusions of actinolite, sphene, iron ore, diopside and epidote. Chlorite is sometimes interstitial.

PETROGENESIS.

(i) The Circulation of Hydrothermal Solutions.

It is evident from the field occurrence and from the petrography that some of the rocks have suffered alteration after having reached a certain stage of contamination. Thus the actinolite-serpentine-epidote assemblage occurs in veins and irregular patches in the actinolite-albiteclinozoisite rock, and the latter has the appearance of having been permeated by serpentine-bearing solutions. From the petrography it is evident that the serpentine replaces albite, and it is possible that the resulting serpentine contains a trace of alumina. Some of the alumina of the albite may join with lime and iron to form epidote, a mineral which is very well developed in

this rock. The hydrothermal solutions would, therefore, appear to have contained magnesia, iron, silica and lime. Syromyatnikov⁽¹²⁾ has shown experimentally that silica and iron may be transferred by water vapours, and he concludes that not only are these oxides transferred in solution, but also there is a molecular migration in the gaseous medium. In the case of the late-hydrothermal stage at Ben Bullen it is likely that water vapour played a very important part.

The vein-like occurrence of the prehnite-bearing assemblage suggests that it has had a similar origin, and in this instance the solutions contained less magnesia and more lime. It is also possible that the initial contamination was brought about by a less magnesian limestone.

The status of the actinolite-chlorite-epidote rock is rather uncertain, but its texture suggests that chlorite represents the hydrothermal alteration of albite, and this suggestion is strengthened by the occurrence of veins and small patches of chlorite in the actinolite-albite-clinozoisite rocks.

The spinel-bearing assemblage contains interstitial calcite and chlorite, and this mode of occurrence suggests deposition from solution, but the genesis of this rather unique type is dealt with more fully on page 153.

These hydrothermally altered rocks represent extreme cases of contamination, and the solutions responsible for their alteration have been derived in part from the magma and in part from the limestone. The rocks are thus allied to metasomatized limestones, but their origin is indirect. Metasomatized limestones are formed by the action of late-magmatic solutions upon solid limestone. These assemblages appear to have arisen by the action of latemagmatic solutions plus limestone solutions upon rocks that have been contaminated by limestone.

(ii) Mineralogical Interpretation of Magnesian Limestone Assimilation.

In the case of the endogenous contact of the nonmagnesian limestones⁽⁸⁾ the field relations are such that each stage in the contamination process can be traced. In this contact-zone, however, the field evidence gives no clue to the degree of contamination, but the petrography indicates that the various assemblages have arisen by gradual increments of magnesia and lime to the quartzmica-diorite magma. It is shown above that these increments have not been brought about by a direct increase in the amount of solid limestone, and that contamination is partly due to the action of late hydrothermal solutions.

Regardless of how these changes have been brought about, it is of interest to trace the mineralogical response to a gradually increasing magnesia content.

As the amount of dolomite present in the magnesian limestones is very variable, further complications arise with regard to this contact-zone.

The first stage in the contamination of the quartz-micadiorite is the formation of a diopside-bearing quartz-diorite in which biotite is not developed. Diopside-plagioclase xenoliths are enclosed in this contaminated diorite, and it is evident that they represent metasomatized limestones.

No true dolomites are known at Ben Bullen, and most of the marbles have a low alumina content. It must be assumed, therefore, that the excess lime has passed out to the magma, and that the plagioclase of the xenoliths has been mechanically derived from the igneous rock. This is supported by the petrography (see page 139).

C. E. Tilley⁽¹³⁾ has shown that masses of pure diopside rock may be formed by the simple addition of silica to dolomite, and it is probable that the diopside of the Ben Bullen xenoliths and of the contaminated rocks enclosing them arose in this way. The excess lime from the magnesian limestone further reacted with the magma and inhibited the formation of biotite.

In the endogenous contact-zone of the non-magnesian limestones the disappearance of hornblende preceded that of biotite, and at a later stage biotite gave place to orthoclase. In this contact-zone, however, hornblende persists for a time, magmatic potash and alumina rapidly decrease and no orthoclase occurs. A little potash and alumina may enter the hornblende molecule, and it is possible that lime combines with part of the potential biotite molecule to form hornblende.

With a further increase in lime and magnesia and a concomitant decrease in potash and alumina, hornblende passes to actinolite, and at the same stage in the contamination process diopside passes to actinolite by the addition of magnesia, silica and water.

At the initial stage of contamination the plagioclase becomes more calcic, but at the stage when hornblende gives place to actinolite the felspar also suffers a change,

and with addition of lime, breaks up into clinozoisite (or epidote) and albite. Later by reaction with magnesia, the albite passes to chlorite according to the following equation :

 $2\{Na[AlSi_{3}O_{8}]\}+5Mgo+4H_{2}O=(OH)_{8}Mg_{5}Al[AlSi_{3}O_{10}]$ $+3SiO_2+Na_2O.$

Finally magnesia displaces alumina and the resulting rock is an actinolite-serpentine-epidote assemblage.

In the chlorite- and serpentine-bearing assemblages the excess lime possibly combines with the displaced alumina to give a further quantity of epidote, which is abundant in these rocks. The prehnite-bearing rocks also indicate addition of lime.

In the following table the entry of magnesia and/or lime from the right is assumed.

		IABLE I.		
Quartz-mica- diorite.	Diopside-bearing Quartz-diorite.	Actinolite- albite- clinozoisite Rock.	Actinolite- chlorite- epidote Rock.	Actinolite Skarn.
Biotite	-→Hornblende	\rightarrow Actinolite —	\longrightarrow Actinolite \longrightarrow	
Hornblende	→Hornblende →Diopside	-→Actinolite -→Diopside	—→Actinolite —- —→Actinolite —-	→Actinolite →Actinolite
Quartz ———	-→Diopside	→Diopside ——	\longrightarrow Actinolite \longrightarrow	→Actinolite
Iron Ore	→Sphene →Iron Ore	-→Sphene		
Andesine ———	→Labradorite	→Clinozoisite –	\longrightarrow Epidote \longrightarrow Chlorite	

(iii) The Genesis of the Exceptional Types.

(1) Diopside-plagioclase-zoisite Rock with White Mica.

The calculated analysis (column III of Table II) suggests that there has been but little selective diffusion during the formation of this contaminated rock, and that it has arisen by the simple addition of magnesian limestone to the quartz-mica-diorite magma.

Brucite-marble occurs abundantly in the Main Quarry and the density (2.64) indicates a mixture of 24% brucite and 76% calcite. Actually there is a little dolomite and iron ore in the rock, so it is probable that calcite is slightly

lower in the actual rock, and this is confirmed by the excess of lime in the calculated analysis.

		1114	I.	II.	III.
$\begin{array}{c} \mathrm{SiO}_2\\ \mathrm{Al}_2\mathrm{O}_3\\ \mathrm{Fe}_2\mathrm{O}_3\\ \mathrm{FeO} \ . \ .\\ \mathrm{MgO}\\ \mathrm{CaO} \ . \ .\\ \mathrm{Na}_2\mathrm{O}\\ \mathrm{K}_2\mathrm{O}\\ \mathrm{TiO}_2\\ \mathrm{P}_2\mathrm{O}_5\\ \mathrm{MnO} \end{array}$	··· ·· ·· ·· ··	··· ··· ··· ··· ···	$55 \cdot 35 \\ 20 \cdot 89 \\ 3 \cdot 62 \\ 5 \cdot 22 \\ 2 \cdot 75 \\ 7 \cdot 45 \\ 2 \cdot 49 \\ 0 \cdot 82 \\ 0 \cdot 90 \\ 0 \cdot 41 \\ 0 \cdot 10$	$ \begin{array}{c} 48 \cdot 84 \\ 18 \cdot 67 \\ 0 \cdot 74 \\ 6 \cdot 63 \\ 5 \cdot 61 \\ 14 \cdot 03 \\ 2 \cdot 55 \\ 1 \cdot 18 \\ 1 \cdot 39 \\ 0 \cdot 24 \\ 0 \cdot 12 \end{array} $	$ \begin{array}{c} 48 \cdot 95 \\ 18 \cdot 45 \\ 3 \cdot 20 \\ 4 \cdot 63 \\ 5 \cdot 68 \\ 14 \cdot 95 \\ 2 \cdot 20 \\ 0 \cdot 73 \\ 0 \cdot 79 \\ 0 \cdot 35 \\ 0 \cdot 07 \end{array} $
			100.00	100.00	100.00

TABLE II.

- I. Quartz-mica-diorite, corrected to 100% with water omitted.
- II. Diopside-zoisite-mica rock corrected to 100% with water omitted.
- III. Calculated contaminated rock consisting of 9 parts of I and 2 parts of $(MgO/CaO = 16 \cdot 56/42 \cdot 56)$ and corrected to 100%. The MgO/CaO ratio is equivalent to 24% brucite and 76% calcite.

From the analyses it would appear that there has been a little selective diffusion of the alkalis, particularly of potash, but it is difficult to interpret this mineralogically, as the conversion of felspar to mica implies an addition of alumina or a subtraction of alkalis.

It is evident that in some cases plagioclase has crystallized from the contaminated magma and has been altered to white mica at a later stage, but in other cases the white mica appears to have crystallized directly. The first stage seems to be the splitting of the actual or potential plagioclase molecule into albite and anorthite, and the subsequent conversion of anorthite into zoisite by the addition of lime and water. The albite molecule then joins with the potential hornblende molecule to give white mica (possibly paragonite and muscovite) and diopside. The resulting rock is, therefore, a diopside-zoisite-white mica assemblage.

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The fundamental mineralogical changes thus appear to have been brought about by the addition of lime, and the added magnesia simply implies a higher diopside content. These changes may be represented by the equations :

- (1) Plagioclase + Water + Lime = Zoisite + Albite.
- (2) Hornblende + Albite + Water + Lime =

White Mica + Diopside.

(2) Pyroxene-spinel-chlorite Rock.

The intersertal texture of this rock would suggest that chlorite (with perhaps some of the calcite) has been derived from hydrothermal solutions, and that the original rock consisted of a granoblastic mosaic of pyroxene and spinel with large areas of calcite against which the other minerals show idioblastic outlines.

It has been shown that the chlorite is a highly ferruginous type and that it contains only small amounts of alumina and magnesia, so it is evident that the hydrothermal solutions carried iron, silica and water. It is therefore necessary to postulate that most of the alumina $(24 \cdot 51 \%)$ was contained in the original rock. It is difficult to see how a spinel-pyroxene assemblage could have arisen by contamination, as the quartz-mica-diorite magma itself contained only about 20% of alumina. It is possible that alumina may have been segregated, but the body of the rock does not suggest crystallization from an aqueous solution or melt.

The association spinel-diopside is characteristic of the high-grade thermal metamorphism of magnesian limestones which contain alumina and a little silica,⁽⁶⁾ and though no such sediments are known at Ben Bullen, the very restricted occurrence of this type suggests that there may have been one seam of this composition.

The assemblage may therefore be regarded as a metasomatized limestone.

(iv) The Absence of Certain Silicate Minerals.

Hatch and Rastall⁽⁷⁾ and du Toit⁽³⁾ have described the endogenous contact of magnesian limestones with granite at Port Shepstone, Natal, and certain differences between the mineral assemblages of this contact-zone and that of Ben Bullen are noteworthy.

At Port Shepstone phlogopite is abundantly developed, and scapolite is also prominent. At Ben Bullen neither K-November 6, 1935. of these minerals occurs. It is evident that halogens were almost lacking in the Ben Bullen magma and the absence of these common lime and magnesian minerals may be ascribed to this deficiency. The absence of vesuvianite from the non-magnesian limestone contact may be similarly accounted for.

(v) Reciprocal Reaction and Prevailing Physical Conditions.

In the case of the endogenous contact of the nonmagnesian limestone tongues of diorite have given rise to definite contaminated zones. In dealing with a single tongue, therefore, it was possible to trace the stages of contamination of that tongue alone. The limestones of this contact, moreover, had a fairly constant composition, and it was obvious that the final stage in the contamination process was the production of a metasomatized limestone, which probably represented a volume-for-volume change. There was thus some basis upon which a series of analyses might be compared.

In the contact-zone of the magnesian limestones, however, it is useless to make a series of analyses. First, the field relations are such that the contaminated rocks cannot be collected about a single igneous mass, and it is uncertain whether two different mineral assemblages represent different admixtures of the same limestone and the same igneous rock. Secondly, the limestones vary in their CaO/MgO ratio. Thirdly, there are further complications brought about by hydrothermal solutions carrying ingredients from the limestones themselves, and it is doubtful whether the interchanges have been volume replacements.

Reciprocal reaction, therefore, cannot be deduced from chemical analyses, but it may be inferred from the mineralogical discussion and from Table I. It is evident that the limestones have contributed lime and magnesia to the igneous rock, and that during the first stages of contamination all the common oxides of the igneous rock have played a part. At a later stage there is a marked diminution of alumina and alkalis, and finally there has been selective diffusion of silica, iron and water from the igneous rock. It is probable that the actinolite-skarn has a small alumina percentage, so the final solution probably carried a little alumina as well.

The physical conditions prevailing during assimilation in this contact-zone are much the same as those of the

non-magnesian limestone contact. It has been pointed out in connection with the latter that the occurrence of microperthite indicates a fairly high temperature.⁽⁴⁾ This mineral occurred in the least contaminated rocks, and it is obvious that high-grade conditions also prevailed at the initial stage of assimilation on the magnesian limestone. The mineralogical response to this was the production of a more basic plagioclase, and splitting of the plagioclase molecule into albite and clinozoisite did not occur until the next stage was reached, when temperatures were obviously lower and water more abundant. The presence of actinolite is also indicative of such conditions.

Pyrites and magnetite occur in the skarn and its associated pegmatite, and these minerals indicate a moderately low temperature—a temperature low enough to inhibit the reaction between quartz and calcite.

The prehnite-, serpentine- and chlorite-bearing rocks probably belong to a slightly later stage, when the temperature was still lower and water vapour was more concentrated.

It is thus evident that contamination took place at low temperatures and in the presence of volatiles, of which water was the most active. Halogens were entirely absent.

(vi) Comparison of the Endogenous Contacts of the Magnesian and Non-Magnesian Limestones.

It has already been pointed out that the field relations are not well defined in the contact of the magnesian limestones, and that matters are further complicated by the variability of the magnesian content, and by the action of hydrothermal solutions. It must also be pointed out that the igneous rock at the contact of the magnesian limestones is a quartz-mica-diorite, whilst that responsible for altering the non-magnesian limestones is a little more basic and contains pyroxene.

In a general way, however, certain similarities are brought out by a comparison of the mineral assemblages of the two contact-zones. Both have arisen under somewhat similar physical conditions. In the one case contamination is brought about by the assimilation of lime, in the other by increasing magnesia and lime; and it is possible to compare the degree of contamination in each case, as is done in Table III.

TABLE III.

Degree of Con- tamination.	Increasing Lime.	Increasing Lime and Magnesia.
I	Diorite Core.	Diopside-bearing Quartz-
	Diopside, plagioclase, orthoclase, iron ore and	diorite. Diopside, plagioclase,
II	Clinozoisite Zone	quartz. Actinolite-albite-
	Diopside, clinozoisite, and albite.	clinozoisite Rock.
III	Prehnite Zone. Diopside, prehnite, clino-	Actinolite-chlorite- epidote Rock.
	zoisite.	(Soda of albite replaced
	by lime.)	by magnesia.)
IV	Garnet Zone. (Sometimes skarn.)	Actinolite Skarn. Actinolite (with some
	Garnet, diopside, wol- lastonite, quartz, calcite.	alumina), quartz, calcite.

SUMMARY.

It has been shown that a mass of limestone of variable magnesia content has been invaded by a quartz-mica-diorite magma, and that a certain amount of assimilation has taken place.

The parent rocks and the contaminated types are described in detail, and it is concluded that all the contaminated rocks may be accounted for by a gradual increase in the amount of MgO and CaO added to the magma.

It is shown, however, that these gradual increments do not take place directly, but that certain rocks have been contaminated first by the assimilation of solid limestone and then by solutions derived from the sedimentary rock as well as from the magma.

Two somewhat exceptional types have been analysed and their genesis is discussed in some detail.

It is pointed out that the absence of certain silicate minerals may be accounted for by the deficiency of halogens in the magma, and it is concluded that the prevailing physical conditions were those of low temperature and

an abundance of water. Reciprocal reaction is believed to have taken place, but some of the primary interchanges have been masked by later reactions.

Finally, the endogenous contact-zones of the magnesian and non-magnesian limestones are compared with regard to the degree of contamination that each assemblage represents.

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