The relationship of orthopyroxene to pigeonite. (With Plate XIII.)

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Introduction.—Interest in the orthopyroxene series is evidenced by the great number of publications on that subject which appeared in the literature. Correlations of the chemical composition and optical properties were attempted by Winchell (1923, 1933), Walls (1935), Henry (1935), and Hess and Phillips (1940). The nomenclature of the series was discussed by Winchell (1923), Alling (1936), and Hess and Phillips (1940). The obscure phenomena exhibited by certain orthopyroxenes in polarized light, attributed by some to twinning, by others to exsolution, was noted by a great many petrologists, of whom are mentioned here Lewis (1908), Holmes and Harwood (1928), Guimarães (1933), Scholtz (1936), Hess and Phillips (1938, 1940), Kuschke (1939), Wager and Deer (1939), Nel (1940), Walker (1940, 1943), Walker and Poldervaart (1940, 1941), Edwards (1942), Henry (1942), and Poldervaart (1944, 1946). Other authors are mentioned by Henry (1942). Attempts to define the stability fields of orthopyroxenes and pigeonites were made by Hess (1941), Walker (1940, 1943), Walker and Poldervaart (1941), Edwards (1942), and Poldervaart (1944, 1946).

The present research partly confirms and extends previously expressed views, and is partly an attempt to summarize the present-day knowledge of this group of rock-forming minerals.

Nomenclature.—It appears most logical to subdivide the three series of solid solutions, formed by the most common groups of rock-forming minerals (i.e., the plagioclase, olivine, and orthopyroxene series) in an analogous manner. The subdivision of the plagioclase series is now generally accepted. Deer and Wager (1939) proposed a similar classification for the olivine series. It is here suggested to subdivide the orthopyroxene series in like manner (text-fig. 1).

The proposed subdivision of the series necessitates a new name to cover the range Of_{70-90} . Since analysed orthopyroxenes within this

range have been described mainly from eulysites, the name 'eulite' is here proposed.

Chemical data.—Two new analyses of magnesian orthopyroxenes, their atomic ratios to 6(0, OH) atoms, and the estimated formulae¹ are given in table I.



TEXT-FIG. 1. Nomenclature of the plagioclase, olivine, and orthopyroxene series. (Molecular percentages.)

The object of the analyses was to test the statements of Hess and Phillips (1938, 1940) regarding the lime content of orthopyroxenes. Hence extreme care was taken to ensure purity in the samples submitted for analysis. In both cases the material was hand-picked and checked repeatedly under the microscope.

The Hanover bronzite forms large 'phenocrysts' (average length 1.5-2.5 mm.) in a medium-grained, ophitic olivine-dolerite (Walker and Poldervaart, 1942). Most of the crystals show a peculiar patchy extinction in polarized light, but some exhibit the 'graphic intergrowth' on an extremely fine scale along their margins (pl. XIII, fig. 1). The Rustenburg norite was obtained from the Schilpadnest mine, Rustenburg, and occurred in the upper part of the Merensky Reef, immediately

¹ Conforming to earlier practice (Henry, 1935; Hess and Phillips, 1940) molecular proportions of MnO were added to those of FeO in calculating the compositions. All other admixtures were neglected.

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TABLE I. Chemical analyses of bronzite. (Analyst, W. H. Herdsman.) Bronzite from Karroo dolerite at Hanover Cane Province

	DI	Unzite II	om Karroo	uolerite at 1	lanove	n, Cape	r rovince.		
	Analysis.		100%.	Atomic ratios.			No. mols. on basis		
	-						6(O,0)H).	
SiO ₂		52.86	53.33	Si		0.889	1.898) 1.996	
TiO ₂		0.26	0.26	Al	••••	0.081	-0.173)98	
Al ₂ O ₃		4 ·08	4 ·12	Fe		0.016	0.034		
Fe ₂ O ₈		1.26	1.27	Fe		0.157	0.336		
FeO		11.23	11.33	Mgʻʻ		0.677	1.444	}1 ∙995	
MnO		0.23	0.23	Ca		0.043	0.092		
MgO		26.82	27.06	Mn"	•••	0.003	0.007		
CaO		2.38	2.40	Ti		0.003	0.007)	
Na ₂ O	•••	trace		0″	•••	2.810			
K ₂ Ō		trace		Formul	a: (Mg,	Fe ^{···} ,Ca	,Mn,Ti,Al)[(Si,Al),O6]	
P_2O_5		nil			Wo	.,En,8.9	Fs ₁₈ .2 or Of	19-1	
$H_2O +$		0.53	100.00						
$H_2O -$	••••	0.07	100.00						
Total		99.72							

Colour, green; pleochroism none; sp. gr. 3.42, $\alpha 1.680$, $\gamma 1.692$, $2V_{\alpha}77^{\circ}$.

	Analysi	8.	100%.	Ato	omic ra	tios.	No. mols 6(O,	. on basis OH).
SiO2		54.26	54.62	Si		0.910	1.936	1.998
TiO ₂	•••	trace		Al		0.030	$0.064 \begin{cases} 0.000 \\ 0.0000 \end{cases}$	062 / - 000 002
Al ₂ O ₃		1.53	1.54	Fe		0.020	0.043	
Fe ₂ O ₃		1.56	1.57	Fe"		0.164	0.349	1.000
FeO		11.71	11.79	Mg"		0.744	1.583	(1.998
MnO		0.43	0.43	Ca		0.004	0.008	ł
MgO		29.68	29.87	Mn		0.006	0.013)
CaO		0.18	0.18	Ti		—	_	
Na ₂ O		nil	_	0″		$2 \cdot 813$		
K₂Ō		nil		Formula:	(Mg,Fe	,Fe'',C	Ca,Mn,Al) [(Si,Al) ₂ O ₆]
P ₂ O ₅		nil			Wo0.41	En _{s1} .1Fs1	8-3 or Of18.	6
H,0+		0.42	100.00					
$H_2O -$	• •••	0.09	100.00					
Total		99·86						

Bronzite from Bushveld norite at Rustenburg, Transvaal.

Colour, brown; pleochroism moderate; sp.gr. 3.40, α 1.679, γ 1.691, 2V $_{\alpha}$ 77°.

below the upper chrome band. A few of the crystals show sporadic exsolution lamellae, but the majority exhibit extremely fine lamellar twinning (pl. XIII, fig. 2) of the type described by Henry (1942) and Scholtz (1936).

Correlation of optical properties and chemical composition.-Textfig. 2 shows the variation in optical properties with chemical composition. The data used are given in table II

Orthopyroxene-pigeonite stability fields.—Various authors have commented on the abnormal behaviour in polarized light of orthopyroxenes within the range Of_{15-35} . In basalts the mineral generally extinguishes quite normally. In dolerites the orthopyroxene may show a peculiar uneven extinction, or more commonly, sets of finely ruled lamellae and irregular blebs of a mineral of different extinction and double refraction than the orthopyroxene host (pl. XIII, figs. 1 and 3). The structure may occur over the entire crystal, or only in its centre, or confined to the margins. Following Walker (1940), the phenomenon is called the



TEXT-FIG. 2. Optical properties and chemical composition of the orthopyroxen^e series. (Molecular percentages.)

'graphic intergrowth', and the patchy extinction, sometimes observed, is regarded as representing the 'graphic intergrowth' on a submicroscopic scale.
 TABLE II. Chemical analyses of orthopyroxenes calculated to 100% and arranged in order of mol.% of orthoferrosilite (Of).

Ref.										
Number	1.	2.	3.	4.	5.	6.	7.	8.	9.	10.
SiO ₂	59 ·8	57.1	53.8	55.9	53.7	54 ·6	53.3	52.0	51.6	53.6
TiO ₂		_	0.1		0.1	_	0.3		0.8	0.3
Al ₂ O ₃	_	0.9	1.7	1.7	4.5	1.5	4·1	3.9	$2 \cdot 2$	2.2
Fe ₂ O ₃		0.4	1.5	1.4	0.3	1.6	1.3	—	4.5	0.8
FeO	0.4	6.4	6.7	7.2	11.6	11.8	11.3	13.3	13-9	15.3
MnO			0.2	0.2	0.2	0.4	0.2	0.6	0.2	0.3
MgO	39.5	34.9	33.2	32.5	$29 \cdot 2$	29.9	$27 \cdot 1$	27.5	$24 \cdot 5$	24.7
CaO	0.3	0.1	$2 \cdot 3$	0.7	0.4	0.2	$2 \cdot 4$	2.0	1.9	2.6
Na ₂ O		0.2		—		_		—	0·4	0.1
K ₂ O	_		0.1	—				—	—	
P ₂ O ₅	-	—		0.1	—	—				—
Cr ₂ O ₃			0.4	0.3		_		0.7	—	0.1
2V	54° (+)	75° (+)	87° (+)	89° (+)	84° (-)	77° (-)	77° (-)	69° (-)	n.d.	66° (-)
α	1.651	1.666	n. d.	1.668	1.675	1.679	1.680	1.681	1.690	n. d.
γ	1.659	1.675	1.677	1.679	1.685	1.691	1.692	1.696	1.700	1.699
γ-α	0.008	0.009	-	0.011	0.010	0.012	0.012	0.012	0.010	—
Sp. gr. Of.	n. d.	3.25	n.d.	n. d.	3.40	3.40	3.42	n. d.	3.42	n. d.
(mol. %)	0.6	9.3	10.4	11.2	18.3	18.6	19-1	21.9	24-2	26 ·0
11.	12.	13.	14.	15.	16.	17.	18.	19.	20.	21.
51.5	52 ·0	53-2	50·0	50.0	50.6	48·2	47.9	47-4	46-4	46 ·5
0.9	0.4	0.2	0.6	0.4	0.3	_	0.1	_	0.2	0.1
0.2	1.7	1.4	3.6	$2 \cdot 8$	1.9	0.7	1.5	0.1	0.3	0.9
0.8	3.2	1.1	—	1.9	$2 \cdot 1$	1.3	0.5	0.3	0.2	0.3
19.5	18.3	18.7	26.7	27.5	29.7	37.8	35.7	42·1	45.0	42.1
0.1	0.4	0-9	0.2	0.6	0.2	2.5	6.3	3-9	1.2	5.1
$25 \cdot 1$	22.2	23.3	16.8	15.6	13.8	7.0	7·1	5.1	5.1	3.2
1.0	1.8	1.2	$1 \cdot 2$	0.9	1.4	2.5	0.9	1.1	1.6	1.5
0 ∙ 4		-	0.1	0.1						_
0.2			0.2	0.2	—	—	_			
—	-		0.3			—	—			
			_	_					_	
63°()	n, d.	60°(-)	53°	55°	51° (-)	70°(–)	68° ()	81° (—)	83° () 83° (+)
1.698	1.702	1.691	n. d.	n. d.	1.715	1.740	1.738	1.750	1.751	1.755
1.713	1.712	1.705	1.730	1.731	1.731	1.757	1.755	1.768	1.769	1.773
0.012	0.010	0.014			0.016	0.017	0.017	0.018	0.018	0.018
n. d.	3.48	n.d.	3.53	3.56	3.60	3.77	3.75	3.83	3.84	3.88
30.2	31.9	31.9	47.4	50.0	54.7	76.2	76.6	83-3	83.4	88.2

Enstatite, Shallowater meteorite (Hess and Phillips, 1940).
 Enstatite, Espedalen, Norway (Washington and Merwin, 1923).
 Enstatite, Stillwater, Montana (Hess and Phillips, 1940).
 Lastatite, Webster, North Carolina (Hess and Phillips, 1940).
 Lastatite, Webster, North Carolina (Hess and Phillips, 1940).
 Lapland (Mikkola and Sahama, 1937).
 Brotzite, Rustenburg, Transvaal (this paper).
 Bronzite, Rustenburg, Montana (Hess and Phillips, 1940).
 Hypersthene, Stillwater, Montana (Hess and Phillips, 1940).
 Hypersthene, Komagataké, Japan (Közu, 1934).
 Hypersthene, Odawara-mati, Japan (Kuno, 1938).
 And 15, Hypersthene, Quilquox, Aberdeenshire (Walls, 1935).
 Hypersthene, Glen Buchat, Aberdeenshire (Henry, 1935).
 Horry, 1935).
 Hypersthene, Tunaberg, Sweden (Sundius, 1932).
 Mansjö, Sweden (Henry, 1935).
 Hypersthene, Tunaberg, Sweden (Sundius, 1932).
 Mansjö, Sweden (Henry, 1935).
 Hypersthene, Tunaberg, Sweden (Sundius, 1932).

In norites the orthopyroxene frequently shows one, two, or three sets of well-developed lamellae, while the irregular blebs are absent. The structure is on a much coarser scale and more regular than the 'graphic intergrowth'. Following Hess and Phillips (1938), it is termed the 'lamellar intergrowth' (pl. XIII, fig. 4).

A great deal of confusion has been caused by the occurrence of fine lamellar twinning in orthopyroxenes (pl. XIII, fig. 2), which is often very similar in appearance to the 'graphic intergrowth'. Twinning of this type was described by Scholtz (1936) and Henry (1942), while it is also exhibited by the Rustenburg bronzite. Henry ascribes the structure to deformation during crystallization.

The patchy extinction, 'graphic intergrowth', or 'lamellar intergrowth' is caused by exsolution of lime. The present author agrees with Hess and Phillips (1938, 1940) that the structure is produced in either of two ways, viz.:

(a) By the inversion of earlier-formed, unstable pigeonite to orthopyroxene upon slow cooling, accompanied by the expulsion of lime from the molecule.

(b) By the exsolution of $CaSiO_3$, dissolved in the orthopyroxene molecule, likewise on further slow cooling.

The relative stabilities of orthopyroxene and pigeonite are rather closely linked under magmatic conditions. Pigeonite appears to be the stable phase at high temperatures, but is unstable at lower temperatures, when orthopyroxene becomes the stable phase (Hess, 1941). Thus, on quick cooling, pigeonite is preserved in a metastable condition, while slow cooling results in the inversion of pigeonite to orthopyroxene with the 'graphic' or 'lamellar' intergrowth.

Pigeonite remains the stable phase at all temperatures in the presence of high volatile concentrations, orthopyroxene becoming unstable under such conditions. This relationship is demonstrated by the following facts:

- (i) Dolerite-pegmatites may contain pigeonite, but there is no record of dolerite-pegmatites in which orthopyroxene occurs.
- (ii) Granophyres, formed in the Karroo by the metasomatism of siltstones by dolerite magma (Walker and Poldervaart, 1942), may contain pigeonite, but orthopyroxene is absent.
- (iii) At Mount Arthur, tholeiites evolved from olivine-dolerites through the distillation of water from large sedimentary xenoliths, contain pigeonite, but no orthopyroxene (Poldervaart, 1946).

If magmatic conditions move towards the stability field of pigeonite, by a gradual concentration of volatile elements, orthopyroxene crystallizes with an increasing amount of dissolved lime, which is again exsolved on cooling. Such conditions prevailed in the Palisade sill (Walker, 1940), the Whin sill (Holmes and Harwood, 1928), the Hangnest sill (Walker and Poldervaart, 1941) and the Sedgewick laccolith (Edwards, 1942). These intrusions all carry pigeonite in their upper portions, while orthopyroxene occurs in the lower parts. The last three intrusions are typical bronzite-dolerites with chilled phases containing phenocrysts of bronzite. From the lower contact upwards the 'graphic intergrowth' first appears along the margins of the orthopyroxene crystals, then covers their entire surface, and finally pigeonite occurs instead of orthopyroxene.

The relations of magnesian orthopyroxene and pigeonite (Mg:Fe ratio 4:1) are further summarized in table III.

Table III.	Conditions	of format	tion of magnesian orthopyroxene and pigeonite.
Temperature of magma (Hess, 1941).	First mineral to crystallize.	Condition of magma.	Rate of cooling.
<1100° Slow rate of crystallization	Normal bronzite	dry wet	$ \begin{cases} \text{slow} \\ \text{fast} \end{cases} \text{Normal bronzite} \\ \begin{cases} \text{slow} \rightarrow \text{Bronzite with graphic} \\ \text{intergrowth} \\ \text{fast} \text{'Frozen' as normal bronzite or bronzite with} \\ \text{graphic intergrowth} \end{cases} $
$\pm 1100^{\circ}$ High rate of crystallization	Bronzite with dissolved lime up to 9% CaSiO _s	dry wet	slow →Bronzite with graphic intergrowth →Normal bronzite fast 'Frozen' as lime-bronzite (unstable) or as bronz- ite with graphic intergrowth slow →Bronzite with graphic intergrowth →Pigeonite fast 'Frozen' as lime-bronzite (unstable) or as bronz- ite with graphic intergrowth
>1100°	Pigeonite	dry wet	$ \begin{cases} {\rm slow} & \rightarrow {\rm Bronzite} \text{ with graphic} \\ {\rm intergrowth} \\ {\rm fast} & `{\rm Frozen' as pigeonite} (unstable) \text{ or as bronzite} \\ {\rm with graphic intergrowth} \\ {\rm slow} \\ {\rm fast} \end{cases} \} {\rm Pigeonite} \end{cases} $

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Abstract.—A new classification of the orthopyroxene series is suggested. Two analyses of magnesian orthopyroxenes are given, and curves are drawn to correlate the chemical composition with the optical properties. The stability fields of orthopyroxenes and pigeonites are discussed in detail.

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EXPLANATION OF PLATE XIII.

(All crossed nicols.)

- FIG. 1. Patchy extinction of bronzite 'phenocryst' in dolerite from Hanover, Cape Province. $\times 15$.
- FIG. 2. Lamellar twinning of bronzite in norite from Rustenburg, Transvaal. $\times 38.$
- FIG. 3. Graphic intergrowth of bronzite in diabase from Palisade, New Jersey. $\times 38.$
- FIG. 4. Lamellar intergrowth of bronzite in norite from Bushveld, Transvaal. $\times 38.$



A. POLDERVAART: ORTHOPYROXLNES.