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*Zoned olivines and their petrogenetic significance.*¹

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I. INTRODUCTION.

THE object of this paper is to demonstrate the widespread occurrence of zoned olivines in igneous rocks, to show that the zoning is continuous, that the composition varies from a magnesium-rich centre to an iron-enriched margin, and that these findings have an important bearing on the question of petrogenesis.

I propose to express the composition of olivine by stating only the percentage of fayalite, as the rock-forming olivines belong mainly to the forsterite-fayalite series. There are, however, a number of names in use for the members of this series. Assuming that the two names forsterite and fayalite are insufficient for the needs of classification, it yet seems to me that all but four of the existing names are superfluous. As the series is isomorphous and as the lines of division are purely arbitrary, I suggest the following decimal classification in molecular percentages:

| | | |
|-------------|--------|-------------|
| Forsterite | | 0Fa— 10Fa |
| Chrysolite | | 10Fa— 50Fa |
| Hortonolite | | 50Fa— 90Fa |
| Fayalite | | 90Fa— 100Fa |

The name hyalosiderite was proposed in 1823 by Walcher for a ferruginous variety of chrysolite. At that time it was considered to be the

¹ Read at the Cambridge meeting of the British Association, August 1938, abstract in Rep. Brit. Assoc. Adv. Sci., 1938, p. 418-419.

most ferruginous variety of olivine. As fayalite was described and named in 1840 by Gmelin and hortonolite in 1869 by Brush, the name hyalosiderite is unnecessary and is besides inaccurate, since the mineral it attempts to describe is neither hyaline nor is it a siderite.

Although zoned olivines have been previously recorded and described their widespread occurrence has never been recognized, and, as a rule, they receive no mention in the text-books.

If we except a few records of zonally altered olivines by F. Zirkel (1), K. Hofmann (2), L. van Werveke (3), and J. J. H. Teall (5, p. 164), the first name demanding notice is that of B. Doss (4), who not only described zoned olivines from Syrian basalts and noted the variation of polarization-colours in the various zones, but also attempted to give an interpretation of this phenomenon as being due to variation in the chemical composition of olivines. His conclusions were that zoned olivines have an iron-rich centre and a magnesium-rich margin. In this he was supported by F. Möhle (10) and W. Schultz (11), who based their conclusions on the zonally disposed alteration products. It will be shown later on that olivines with a magnesium-rich margin are found in nature, but only as exceptions to the general rule.

The discovery of the relation existing between the optical properties and the chemical composition of olivines by S. L. Penfield and E. H. Forbes (6, 7) and the subsequent additional data provided by M. Stark (12), H. Backlund (14), and N. H. Magnusson (19) have provided mineralogists with a useful optical method for the study of olivines. In their final form the curves showing the relation between the optical properties and the chemical composition of forsterite-fayalite series are given by A. N. Winchell (31, p. 191).

A. Sigmund (8) was the first to apply the optical method to the study of zoned olivines from Styria. He found that (+) $2V$ of the outer zone is by 7° greater than that of the centre and that the outer zone shows higher polarization-colours than the centre. He therefore concluded that the outer zone is more ferruginous than the centre. F. Becke (9) also found that in the olivine from Predazzo melaphyre the difference of $2V$ between the centre and the margin was 7° , and in the olivine from the quartz-basalt of Cinder Cone, California, the difference was 5° , and that the outer zone in all cases was more ferruginous than the centre. M. Stark (13) gave a detailed account of zoned olivines from the basalts of the island of Ustica (NW. of Palermo, Sicily). In one of the crystals described by him the calculated amount of fayalite in the centre was $19\frac{1}{2}\%$ and in the margin $34\frac{1}{2}\%$. H. Michel (17) described some zoned

artificial fayalites in which the centre was less ferruginous than the margin. H. Backlund (18, 20) described a zoned olivine with 19Fa in the centre and 36Fa in the margin from a nepheline-basalt (onkilonite) from Vilkitzky Island, Arctic Ocean. Recently zoned olivines have been found to be of frequent occurrence in the Siberian traps (dolerites and basalts) which probably belong to the Arctic igneous province (35, 40, 41). In a further description of zoned olivines, this time from the rocks of Etna and Bohemia, M. Stark (23) made the first attempt to find a relation between the composition of the olivine and the composition of its parent rock. In this he was followed by F. Rinkel (25) and T. F. W. Barth (30). The last author also described a zoned olivine with 18Fa in the centre and 25Fa in the margin. Zoned olivines have also been described by W. Campbell Smith and L. J. Chubb (26) from Austral Islands, Pacific Ocean, by P. Esenwein (28) from the Azores, by O. H. Ödman (29) from Mt. Elgon, East Africa, by E. Jérémime (36) from Lake Rudolf, East Africa, by H. Kuno (38, 39) from Kurile Islands, Japan, and by L. Jugovics and A. Marchet (42) from Ságberg, Hungary.

A theoretical explanation of zoning in olivines was first given by C. Doelter who, in his 'Handbuch der Mineralchemie' (15), attributed this to the progressive enrichment of the growing crystals in a more fusible iron orthosilicate. J. H. L. Vogt (21) pursued this theory further and tried to explain zoning on the basis of a tentative thermal equilibrium diagram for the binary system forsterite-fayalite which he assigned to type I of Roozeboom, but it is only recently that N. L. Bowen and J. F. Schairer (37) have succeeded in establishing this diagram on experimental data.

II. VARIOUS METHODS OF OBSERVING AND MEASURING ZONED OLIVINES.

Zonality in olivines can be ascertained by the following four methods:

(1) extinction-angles, (2) degree of birefringence, (3) indices of refraction, (4) optic axial angles.

(1) The first method is very suitable for spotting zoned olivines in thin sections, but does not provide, as yet, an easy method to determine the composition of zonal components. Olivine, like all the orthorhombic crystals, gives straight extinction (as measured from the pinakoidal cleavages) only in sections perpendicular to the principal planes of the optical ellipsoid. The angle of extinction in any other section can easily be determined graphically on a stereographic projection with the help of the law of Biot, which states that the vibration-direction in any section of a biaxial crystal is given by the bisectors of the angle between the optic axes as projected on the plane of the section. To illustrate the

variation of the extinction-angle in a zoned olivine a theoretical case is given in which the centre has $2V = (+) 80^\circ$ and the margin $2V = (-) 80^\circ$. Fig. 1a shows the curves of equal extinction measured from the trace of the dominant cleavage (010). The curves in fig. 1 b are

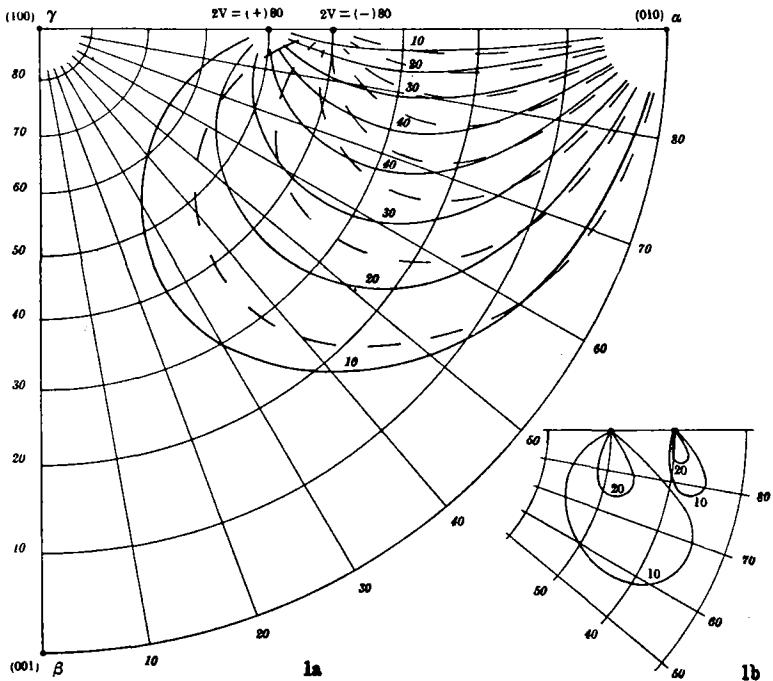


FIG. 1 a. Curves of equal extinction-angles from the cleavage (010) in an orthorhombic crystal for optic axial angles ($2V$) of $(+) 80^\circ$ and $(-) 80^\circ$.

FIG. 1 b. Curves of equal differences of extinction-angles for same.

derived from the curves of equal extinction and represent the values of equal differences between the extinction of the centre and that of the margin. It is obvious, therefore, that we should find the greatest contrast of extinction-angles in the vicinity of the emergence of the optic axes, e.g. in the region of low birefringence. This is supported by actual observation, namely, that one can easily spot a zoned olivine in a thin section of rock by observing the variation of extinction-angle in grains showing grey polarization-colours. I found this method infallible, but it is quite obvious that when the difference in the composition of different zones is small, the movement of the extinction-shadow is slight.

The determination of the optic axial angle from the angle of extinction is theoretically possible if the correct orientation of the section is known. It involves complicated calculations or the use of specially prepared diagrams, and so this method, as applied to the determination of the optic axial angle, can hardly be advised.

(2) The variation in polarization-colours in different zones of markedly zoned olivines is a very striking phenomenon, but only olivines having a considerable zonal range show it. In the forsterite-fayalite series we get the following values for birefringence and polarization-colours in the principal planes of the optical ellipsoid in sections 0.02 mm. thick:

| Plane. | | Forsterite. | Fayalite. |
|---------------|--------|------------------------|--------------------------|
| $\perp\alpha$ | | 0.020 Orange I | 0.009 Blue-grey I |
| $\perp\beta$ | | 0.035 Greenish-blue II | 0.050 Red II |
| $\perp\gamma$ | | 0.015 Yellow I | 0.041 Yellowish-green II |

In olivines with normal zoning, e.g. magnesia-rich centre and iron-rich margin, the colour will rise from the centre to the periphery in sections perpendicular to β and γ , and fall in sections perpendicular to α . This means that in the majority of sections the colours will rise from the centre to the periphery, but it is important to determine the orientation of the section before drawing any conclusion. Great difference in colour can only be expected in the case of marked compositional difference between the centre and the periphery. In the Irish Tertiary dolerites and basalts, for example, olivine varies on the average from 20Fa in the centre to 40Fa at the margin. In this case the greatest variation in birefringence and polarization-colour will be shown in sections perpendicular to γ with $\beta - \alpha$ 0.019 in the centre and 0.023 at the margin. Thus in sections 0.02 mm. thick the colour will vary from orange I in the centre to orange-red I at the margin—a hardly perceptible variation. In thicker sections naturally the variation of colour will be more striking, but, on the whole, olivines from Irish or Scottish Tertiary igneous rocks do not show colour variation in a convincing way. On the other hand, olivines from certain Scottish essexites and from a luscladite from Rapa, Austral Islands, show a striking variation of colour. These rocks will be discussed in the next part of this paper. One may conclude, therefore, that the observation of polarization-colours is only useful for spotting zoned olivines of a wide compositional range. The measurement of birefringence in orientated sections may, however, provide a help in the determination of the composition.

(3) The refractive indices of the members of the forsterite-fayalite series exhibit a continuous variation with the composition, rising with

the increase of iron in the mineral. The measurement of refractive indices in orientated sections of zoned olivines will no doubt give an approximate composition of the various zones, but such a procedure is cumbersome and time-robbing. An interesting effect has been observed by placing a drop of liquid of a suitable refractive index over an uncovered section of a zoned olivine fractured across the middle. On raising the objective the Becke line moved inwards into the mineral in the centre and outwards at the margins, and assumed the form of a sine curve.

(4) The optic axial angle, like the refractive indices, varies with the composition in the forsterite-fayalite series. Compared with the other methods, the determination of the optic axial angle presents the simplest and the surest means of arriving at the chemical composition of olivines. We do not know as yet how far the presence of other components affects the values of the optic axial angle, but their amount in the rock-forming olivines is very small and on the whole we can rely on the mean value curve given by A. N. Winchell (31, p. 191).

The apparent optic axial angle ($2H$) can easily be measured on the Fedorov universal stage. The connexion between the apparent ($2H$) and the true optic axial angle ($2V$) is given by the equation $\sin H = \beta \sin V/n$, in which β is the mean index of refraction of the mineral and n is the index of refraction of the glass hemispheres of the Fedorov stage. The obvious difficulty confronting us in this work is that of finding the mean index of refraction of the mineral actually measured on the Fedorov stage, especially when this mineral is a zoned one. To overcome this difficulty I have calculated the values of $2H$ for all the values of $2V$ and β as given on Winchell's diagram. Table I gives the results of this calculation made for Fedorov stage glass hemispheres of refractive index 1.649. This table enables us to read the value of $2V$ and the corresponding value of fayalite percentage from the measured value of $2H$, and provides us with the basis for all subsequent calculations. One can also trace the curve of $2H$ above the curve of $2V$ on the diagram of Winchell, and use the curve instead of the table.

The results obtained during the present work seem to confirm the data provided by Winchell. A number of olivine crystals whose optic axial angles were measured on the Fedorov stage were also tested for refractive indices and birefringences and no important discrepancy was noted, except in the case of fayalite, which will be discussed later. In my opinion, the optic axial angle gives the best index of the composition of olivine, and this method is far simpler and more accurate than any of the other methods.

The majority of olivine grains were measured from the emergence of both optic axes, but when only one axis was available, an accurate determination of an additional principal plane of the optical ellipsoid allowed this angle to be derived by construction on the stereographic projection. The determination of the optic sign of the direction measured is essential in all this work.

TABLE I. Apparent ($2H\alpha$) and true ($2V\alpha$) optic axial angles for forsterite-fayalite series, together with the estimated percentages of fayalite, for Fedorov stage hemispheres of refractive index 1.649.

| Fayalite % | | | | Fayalite % | | | |
|--------------|--------------|------|---------|--------------|--------------|------|---------|
| $2H\alpha$. | $2V\alpha$. | Mol. | Weight. | $2H\alpha$. | $2V\alpha$. | Mol. | Weight. |
| 94 | 94 | 0 | 0 | 73 | 67 | 64 | 72 |
| 93 | 92 | 8 | 11 | 72 | 65½ | 66 | 74 |
| 92 | 90 | 13 | 18 | 71 | 64½ | 68 | 75 |
| 91 | 88½ | 17 | 23 | 70 | 63½ | 70 | 77 |
| 90 | 87½ | 20 | 27 | 69 | 62½ | 72 | 79 |
| 89 | 86½ | 23 | 30 | 68 | 61½ | 74 | 80 |
| 88 | 84½ | 27 | 35 | 67 | 60½ | 76 | 82 |
| 87 | 83 | 30 | 38 | 66 | 59½ | 78 | 84 |
| 86 | 82 | 33 | 42 | 65 | 58½ | 80 | 85 |
| 85 | 81 | 35 | 44 | 64 | 57½ | 81 | 86 |
| 84 | 79½ | 38 | 47 | 63 | 56½ | 83 | 87 |
| 83 | 78½ | 41 | 50 | 62 | 55½ | 85 | 89 |
| 82 | 77 | 44 | 53 | 61 | 54 | 87 | 91 |
| 81 | 75½ | 46 | 55 | 60 | 53 | 89 | 92 |
| 80 | 74½ | 48 | 57 | 59 | 52 | 91 | 94 |
| 79 | 73½ | 51 | 60 | 58 | 51 | 93 | 95 |
| 78 | 72½ | 53 | 62 | 57 | 50 | 95 | 96 |
| 77 | 71 | 56 | 65 | 56 | 49 | 96 | 97 |
| 76 | 70 | 58 | 67 | 55 | 48 | 98 | 98 |
| 75 | 68½ | 60 | 68 | 54 | 47 | 100 | 100 |
| 74 | 67½ | 62 | 70 | | | | |

The method of determination of the apparent optic axial angle is as follows: the β -direction of the optical ellipsoid of the crystal is brought into coincidence with the K -axis of the stage, the stage placed in a diagonal position and slowly rotated around the K -axis. The position of the optic axes in zoned olivines will be marked by shadows moving from the centre of the crystal to the periphery, or vice versa, according to the position of α - and γ -axes. First, the maximum extinction of the central portion of the crystal is noted and the angle (k) recorded, then the stage is slowly rotated around the K -axis, the moving shadow fixed and the angle recorded for any desired position within the crystal. The difficulty of fixing the maximum shadow on the actual margin of the grain is considerable, and I believe that all the values in this paper relating to the composition of the margin are slightly underestimated. A further

complication is due to the fact that quite a number of olivine crystals have their outer zone replaced wholly or in part by alteration products (serpentine, iddingsite, chlorophaeite, &c.), and these obscure the vision for measurement purposes. In respect of the central parts of olivine crystals, the measurements again fall short of true values, but in a direction opposite to those of the margin. This is due to the fact that only a few sections pass through the true centre of the grain, but in this case the probable error is not very considerable. We may take it, therefore, that the measured compositional range of zoned olivines is somewhat below its true value.

III. OPTICAL DATA.

I have tried for the purpose of this section to determine within the compass of one single olivine crystal the difference in composition from zone to zone and to do this by the measurement of certain optical values along specific distances within the crystal. The olivines selected for these determinations are strongly zoned ones: (1) from luscladite, Rapa, Austral Islands, described by W. Campbell Smith and L. J. Chubb (26); and (2) from essexite, Crawfordjohn, Lanarkshire.

(1) Olivine from Rapa luscladite.—The measurement of the optic axial angle and the extinction-angle from the trace of (010) cleavage, at regular intervals from the centre to the margin, gave the following results:

| | Distance in mm. | 2H. | Angle of extinction. |
|---------------|-----------------|-----|----------------------|
| Centre | 0.0 | 90° | 15° |
| | 0.1 | 90 | 15½ |
| | 0.2 | 89 | 16 |
| | 0.3 | 87 | 17 |
| | 0.4 | 83 | 20 |
| | 0.5 | 77 | 24 |
| Margin | 0.6 | 70 | 29 |

The determination of birefringence in this section cannot be made with any degree of accuracy, but two orientated olivine grains from the same rock gave the following values:

| | $\gamma - \alpha$. | $\gamma - \beta$. |
|---------------|---------------------|--------------------|
| Centre | 0.039 | 0.016 |
| Margin | 0.049 | 0.012 |

(2) Olivine from Crawfordjohn essexite.—The most spectacular example of a zoned olivine was found in essexite, Crawfordjohn, Lanarkshire (fig. 2). Unfortunately, the section is mounted on a large-sized glass slide and thus not suitable for measurement on the Fedorov stage. A further difficulty is that the phenocryst of olivine is trisected by cracks,

but this defect has been remedied on the photograph. The pole of the section is inclined to the c -axis of the crystal at approximately 20° . The dominant cleavage (010) is parallel to the shorter diagonal of the rhomb, while the less prominent cleavage (100) is parallel to the longer diagonal.

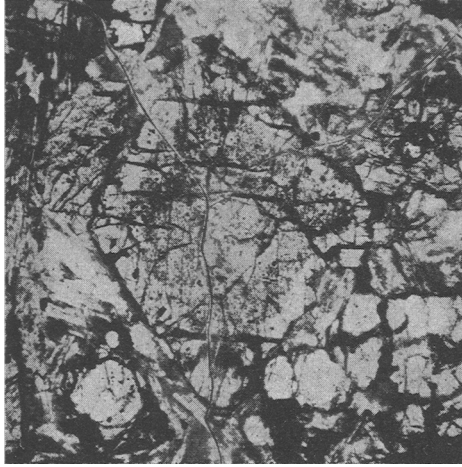


FIG. 2. Zoned olivine in essexite, Crawfordjohn, Lanarkshire. Ordinary light. Magnification 25.

The crystal faces cut by the section belong to the prism zone. They are (120) and (110), and their unequal development explains the asymmetry of the rhombic section. The inner portion of the crystal is full of inclusions of iron-ore and orthite, but the outer zone, some 0.2 mm. thick, is free from inclusions. The polarization-colour of the inner part is blue and that of the outer zone yellowish-green with a narrow bluish-green transition zone. The following values of the birefringence ($\gamma' - \alpha'$) were obtained by means of Berek's compensator:

| | Distance in mm. | $\gamma' - \alpha'$. |
|---------------|-----------------|-----------------------|
| Centre | 0.0 | 0.027 |
| | 0.1 | 0.027 |
| | 0.2 | 0.027 |
| | 0.3 | 0.027 |
| | 0.4 | 0.028 |
| | 0.5 | 0.029 |
| | 0.6 | 0.030 |
| | 0.7 | 0.034 |
| Margin | 0.8 | 0.041 |

Thus the measurement of the optic axial angle, birefringence, and angle of extinction along measured distances in zoned olivines shows that

the variation of these values, and therefore the variation in composition, is roughly proportional to the cube of the distance from the centre of the crystal. When plotted on diagrams (not given in this paper) with x -axis corresponding to the distance from the centre and y -axis to the difference of $2H$, angle of extinction, or birefringence, the curves obtained show a good approximation to the cubic curve $y = x^3$. This means that the change in composition in successive zones is proportional to the volumes occupied by these zones, in other words, the change of composition per unit volume is a constant value in a given olivine crystal. The average composition of a zoned olivine, therefore, will be equal to the arithmetic mean of the composition of the central part and that of the margin.

The birefringence was also measured in some other sections, mainly for the purpose of checking the validity of Winchell's curves. A section of olivine from Crawfordjohn essexite perpendicular to α gave: centre $\gamma-\beta$ 0.017, $2H$ $86\frac{1}{2}^\circ$; margin $\gamma-\beta$ 0.016, $2H$ $81\frac{1}{2}^\circ$. While another perpendicular to β gave: centre $\gamma-\alpha$ 0.037, margin $\gamma-\alpha$ 0.041.

The birefringence $\gamma-\alpha$ was also measured for the olivine from the three varieties of dolerite of Fair Head, Co. Antrim, which will be discussed later. The values obtained were as follows:

| | Centre. | Margin. |
|---|---------|---------|
| Olivine from glomeroporphyritic dolerite | 0.034 | 0.038 |
| „ „ olivine-dolerite | 0.038 | 0.042 |
| „ „ dolerite-pegmatite | 0.043 | 0.047 |

All these results seem to show that we get a fairly good agreement between the values for birefringence and the values for the optical axial angle as given in Winchell's diagram.

The values for the optic axial angle were determined for 88 olivine crystals from various igneous rocks (table II). For convenience, the results are translated into fayalite molecular percentages, but any one interested in the actual angles can easily obtain them by referring to table I. The 88 olivines given in table II are further arranged in 13 groups according to the nature, age, and locality of the rocks, and the averages of these groups are given in table III, together with the difference between the composition of the centre and that of the margin, and also the average of the two values.

The olivines measured were not chosen on account of their zonality, but for their orientation in sections suitable for measurement on the Fedorov stage. Considering the variability of composition shown by olivines, even those belonging to one and the same rock, it may be

TABLE II. Composition of zoned olivines calculated from the optic axial angle.

| Group. No. | Name of rock and locality. | Fayalite Centre. | Mol. % Margin. | |
|---|--|---|----------------|----|
| I | 1. Peridotite inclusion in basalt, Caltor Hill, Derbyshire. 2. | 8 | 8 | |
| | | 13 | 13 | |
| II | 3. Olivine-dolerite. Wooden Hill, Jedburgh, Roxburghshire. 4. | 33 | 34 | |
| | | 35 | 44 | |
| | 5. Olivine-dolerite, Peniel Heugh, Jedburgh, Roxburghshire. 6. | 44 | 53 | |
| | | 20 | 30 | |
| | 7. Olivine-dolerite, Rutherford, Kelso, Roxburghshire. 8. | 20 | 23 | |
| | | 33 | 34 | |
| | 9. Olivine-dolerite, Duns, Berwickshire. 10. | 20 | 21 | |
| | | 20 | 35 | |
| | 11. Olivine-dolerite, Cottonshope, Northumberland. 12. | 38 | 48 | |
| | | 38 | 48 | |
| | 13. Olivine-dolerite, Cottonshope, Northumberland. 14. | 35 | 53 | |
| | | 44 | 51 | |
| | III | 15. Olivine-basalt (Dunsapie type), Queenscairn, Stichill, Roxburghshire. 16. Olivine-basalt (Dunsapie type), Lurdenlaw, Kelso, Roxburghshire. | 0 | 17 |
| | | | 38 | 53 |
| 17. Olivine-basalt (Dunsapie type), East Softlaw, Kelso. 18. | | 53 | 54 | |
| | | 27 | 30 | |
| 19. | | 48 | 50 | |
| IV | 20. Teschenite, Spalefield, Anstruther, Fife. 21. 22. | 13 | 20 | |
| | | 13 | 20 | |
| | | 15 | 28 | |
| | 23. Teschenite, Gosford Bay, East Lothian. 24. | 18 | 25 | |
| | | 23 | 38 | |
| | 25. Teschenite, Gosford Bay, East Lothian. 26. | 13 | 33 | |
| | | 20 | 38 | |
| | 27. Teschenite, Gosford Bay, East Lothian. 28. | 20 | 44 | |
| | | 27 | 48 | |
| 29. | 41 | 58 | | |
| V | 30. Essexite, Crawfordjohn, Abington station, Lanarkshire. 31. 32. 33. 34. 35. 36. | 13 | 38 | |
| | | 32 | 45 | |
| | | 33 | 58 | |
| | | 38 | 52 | |
| | | 38 | 53 | |
| | | 38 | 53 | |
| VI | 37. Glomeroporphyritic dolerite, Fair Head sill, Ballycastle, Co. Antrim. 38. 39. 40. 41. 42. | 0 | 20 | |
| | | 8 | 35 | |
| | | 20 | 20 | |
| | | 23 | 35 | |
| | | 27 | 27 | |
| | | 27 | 56 | |

| Group. No. | Name of rock and locality. | Fayalite Mol. % | |
|------------|---|-----------------|---------------|
| | | Centre. | Margin. |
| VII | Olivine-dolerite, Fair Head sill. | 43. | 27 44 |
| | | 44. | 30 56 |
| | | 45. | 38 66 |
| VIII | Dolerite-pegmatite, Fair Head sill. | 46. | 62 74 |
| | | 47. | 64 80 |
| | | 48. | 64 83 |
| IX | Olivine-dolerite, Portrush sill, Co. Antrim. | 49. | 8 38 |
| | | 50. | 20 33 |
| | Olivine-dolerite, Falbane dike, Ballycastle, Co. Antrim. | 51. | 13 27 |
| | | 52. | 13 33 |
| | Olivine-dolerite, North Star dike, Ballycastle, Co. Antrim. | 53. | 13 44 |
| | | 54. | 20 38 |
| | | 55. | 20 53 |
| 56. | 27 44 | | |
| 57. | 33 53 | | |
| X | Olivine-basalt (lower plateau lava), The Gobbins, Island Magee, Co. Antrim. | 58. | 13 33 |
| | | 59. | 13 41 |
| | | 60. | 17 30 |
| | Olivine-basalt (lower plateau lava), Clogh, Ballymena, Co. Antrim. | 61. | 20 33 |
| | | 62. | 0 23 |
| 63. | 35 46 | | |
| XI | Olivine-basalt, Madras, Oregon, U.S.A. | 64. | 33 44 |
| | | 65. | 33 53 |
| | | 66. | 35 51 |
| | Olivine-basalt, Pocatello, Idaho, U.S.A. | 67. | 13 38 |
| | | 68. | 27 34 |
| | Olivine-quartz-basalt, Cinder Cone, California, U.S.A. | 69. | 30 41 |
| | | 70. | 18 42 |
| | | 71. | 23 29 |
| | | 72. | 23 41 |
| | | 73. | 27 33 |
| 74. | 35 46 | | |
| XII | Luscladite, Tapui Islet, Ahurei Bay, Rapa, Austral Islands, Pacific Ocean. | 75. | 20 70 |
| | | 76. | 33 81 |
| | | 77. | 33 85 |
| | | 78. | 44 87 |
| | | 79. | 44 89 |
| | | 80. | 48 85 |
| | | 81. | 48 87 |
| | | 82. | 48 89 |
| | | 83. | 53 89 |
| | | 84. | 58 96 |
| 85. | 58 96 | | |
| XIII | Fayalite-quartz-porphry, Kудару Hills, Nigeria. | 86. | 100 100 |
| | | 87. | 100 100 |
| | | 88. | 100(?) 100(?) |

objected that the sampling was insufficient and that more measurements should have been made. Although it is true that a larger number of

measurements would increase the accuracy of the results, the time taken by these measurements would prove excessive (the measurement and plotting of each grain takes from one to two hours) while the additional data would probably alter the average but slightly. In order to test this I made 25 additional measurements of the olivine from the Cottonshope olivine-dolerite. The result of these additional measurements shows a total range of 23Fa to 53Fa and an average range of 33Fa to 46Fa, while the average of the tabulated data (table II, nos. 10-14) gives a total range of 20Fa to 53Fa and an average range of 35Fa to 47Fa. The difference is negligible if we take into account the probable error of measurement.

TABLE III. Calculated average composition of zoned olivines in different rock-types.

| | | Mol. % of fayalite. | | | | |
|--|--|---------------------|-----|---------|----------|--|
| | | Centre. Margin. | | Differ- | Average. | |
| Group. | Rock-type. | | | ence. | | |
| British Carboni- ferous- Permian rocks | I Peridotite | 10½ | 10½ | — | 10½ | |
| | II Olivine-dolerite | 31½ | 39½ | 8 | 35½ | |
| | III Olivine-basalt | 33 | 41 | 8 | 37 | |
| | IV Teschenite | 20 | 35 | 15 | 27½ | |
| | V Essexite | 33½ | 51 | 17½ | 42 | |
| Irish Tertiary rocks | VI Glomeroporphyritic dolerite, Fair Head | 17½ | 32 | 14½ | 25 | |
| | VII Olivine-dolerite, Fair Head | 31½ | 55½ | 24 | 43½ | |
| | VIII Dolerite-pegmatite, Fair Head | 63½ | 79 | 15½ | 71 | |
| | IX Olivine-dolerite | 18½ | 40½ | 22 | 29½ | |
| | X Olivine-basalt | 16½ | 34½ | 18 | 25½ | |
| | XI Olivine-basalt, Western States, U.S.A. | 27 | 41 | 14 | 34 | |
| | XII Luscladite, Rapa, Austral Islands | 44½ | 87 | 42½ | 65½ | |
| | XIII Fayalite-quartz-porphry, Kudaru Hills, Nigeria | 100 | 100 | — | 100 | |

Although individual olivines found in the same or similar rocks undoubtedly vary in composition, this variability has a definite range. On the other hand, in olivines drawn from different rock-types the range and average composition differ from type to type. Compare, for example, the three phases of the Fair Head sill (table III, averages VI, VII, and VIII) for dissimilar ranges; and averages II, III and IX, X, XI for similar rock-types. Certainly one must realize that the measured olivines are either phenocrysts or larger grains in the groundmass, and that small grains and ophitic olivines of late crystallization could not be measured. The probable effect of this selection is to lower the range on the fayalite side. Ophitic olivines enclosing feldspar laths are extremely common among the Irish Tertiary

basalts and dolerites and they are invariably richer in iron than the idiomorphic phenocrysts. On the other hand, an analysis of olivine extracted from a rock shows a higher percentage of magnesia than an analysis of the same olivine calculated from the analysis of the rock. This may be partly due to the removal of the highly ferruginous iddingsite borders during the process of extraction of the mineral from the rock, as I have already suggested (34, p. 506), but it may be also due to the removal of the outer ferruginous border and also to the selection of larger crystals for analysis.

The examination of tables II and III shows that the rocks selected vary from ultrabasic to acid. The ultrabasic end is represented only by a peridotite inclusion from a basalt. The olivine in this inclusion is very rich in magnesia. This agrees with a previously published analysis of this olivine (27, p. 713) which, on recalculation, gives 14Fa. Examination of other peridotites confirms the impression that, on the whole, the olivine in them is rich in magnesia, but apparently there are peridotites, like those of the Bushveld Complex in South Africa, in which the olivine is more ferruginous. In dolerites and basalts the average content of fayalite is much higher and varies between 25 and 40. In more alkalic rocks, such as essexite and luscladite, the fayalite content is still higher and the zonality more marked. Unfortunately, in typical teschenites the olivine is too decomposed for the purpose of optical determination. The teschenites given in table II have more affinities with the crinanites, and their olivines are comparable with those of dolerites and basalts. Olivines from Carboniferous dolerites and basalts are not strongly zoned, although these rocks are more alkaline than those from the Irish Tertiary in which good zoning is common. Columbia River plateau basalts, as well as others from the Western States of America, are very similar to the Irish Tertiary basalts and possess well-zoned olivines. In the Siberian basalts and dolerites the olivine is conspicuously zoned (35, 40, 41), and it would be interesting to find if the olivines from the North Atlantic region are zoned and to what degree. The greatest range of zoning is shown by the olivine from luscladite of Rapa, Austral Islands. This olivine is also ferruginous (table III, average XII). Three olivine grains from a fayalite-quartz-porphry of Kudaru Hills, Nigeria, described by A. D. N. Bain (33) correspond to pure fayalite. Two of them gave $2H\ 54^\circ$, a figure which, according to table I, corresponds to 100Fa; but one grain gave $2H\ 50^\circ$. The same low angle has been obtained in a fayalite from a slag. This may be due to the presence of some other constituents or to the imperfection of the olivine diagram.

The composition of zoned olivines reduced to six averages is represented graphically in fig. 3. The range of zoned olivine in each average is represented by an arrow drawn from the point corresponding to the composition of the centre to that corresponding to the composition of

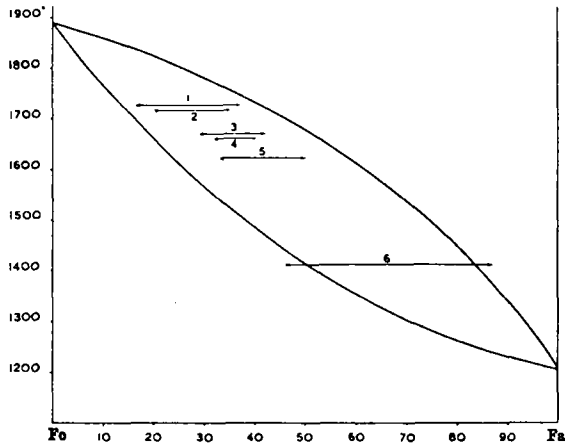


FIG. 3. Average compositional ranges of zoned olivines from different regions plotted against the thermal equilibrium diagram for the binary system forsterite-fayalite. 1, Irish Tertiary dolerites and basalts; 2, British Carboniferous-Permian teschenites; 3, Basalts of the Western States of America; 4, British Carboniferous-Permian dolerites and basalts; 5, British Carboniferous-Permian essexite; 6, Luscladite, Rapa, Austral Islands.

the margin. These arrows are plotted against the thermal diagram for the forsterite-fayalite series as given by N. L. Bowen and J. F. Schairer (37). With the exception of the Rapa luscladite, all the average ranges of zoned olivines fall within the boundaries of the liquidus and solidus curves. That the zonal ranges do not stretch right across the theoretical range marked by the curves is what one would expect, considering the probability of error in measurement and the possibilities of incomplete equilibria and resorption of crystals formed at an earlier stage of crystallization. In spite of these shortcomings, the present investigation fully supports the experimental results obtained by Bowen and Schairer and provides an illustration of the crystallization process as outlined by them. The crystallization of olivines in igneous rocks on the whole follows the course from forsterite to fayalite, and both the individual crystals and the successive crops of crystals tend to become more ferruginous

as the crystallization proceeds. This trend may be called the normal course of crystallization and the consequent zoning—the normal zoning. There are, however, cases of oscillatory and reverse zoning, in which the normal zoning is followed by a reversal to an early magnesium-rich phase. Only a few cases of reverse zoning were noted during the present work, and unfortunately the crystals were not suitable for measurement. It is interesting to note, however, that in the case of reverse zoning the outermost magnesium-rich zone is sharply separated from the inner core. Reverse zoning was the one which was observed by B. Doss (4), F. Möhle (10), and W. Schultz (11), who concluded that it represented the common type of zoning. It was left to A. Sigmund (8) to demonstrate the significance of the normal zoning.

IV. PETROGENETIC SIGNIFICANCE OF ZONED OLIVINES.

Rock-forming olivines belong mainly to the forsterite-fayalite series. Statistical study shows that out of 238 analysed olivines from igneous rocks and meteorites 195, or 82%, contain more than 90% of $Mg_2SiO_4 + Fe_2SiO_4$. It is only olivines from slags that are rich in Ca_2SiO_4 and Mn_2SiO_4 . A frequency-distribution curve constructed on the basis of 257 analyses of olivines (including 19 analyses of olivines from slags) is given in fig. 4. In my opinion this frequency-distribution curve does not correspond to the real frequency-distribution of olivines in nature. This assumption is based on the following argument. A rough estimate indicates that basic rocks with an average olivine content of 15% form about one-third of the total of the igneous rocks of the earth's crust; and although olivine-rich ultrabasic rocks are extremely rich in olivine their relative abundance is negligible as compared with the basic rocks, being about 0.5% of the total of the igneous rocks. The amount of olivine present in the intermediate and acid rocks is far too small to be of any account. Therefore the mode of the frequency-distribution curve should fall at the average composition of olivines from the basic rocks. The average composition of olivine from dolerites and basalts as computed from their optical data (table III) is about 30Fa (mol.) or 38Fa (weight). On the other hand, the mode of the frequency curve (fig. 4) corresponds to 15Fa (weight). In my opinion this mode is lowered through the unconscious selection of olivines analysed, the majority of which are olivines from peridotites and phenocrysts from basalts, both of them exceptionally rich in magnesia. Unconscious selection is probably likewise responsible for the rise of the curve at the fayalite end, as mineralogists are somewhat attracted by rare minerals which they are eager to

analyse on every possible occasion. Actually, fayalite is found only occasionally and in minute quantities in alkalic intermediate and acid rocks. It is the character of the distribution of analyses that makes N. H. Magnusson (19) suggest an immiscibility gap between hortonolite

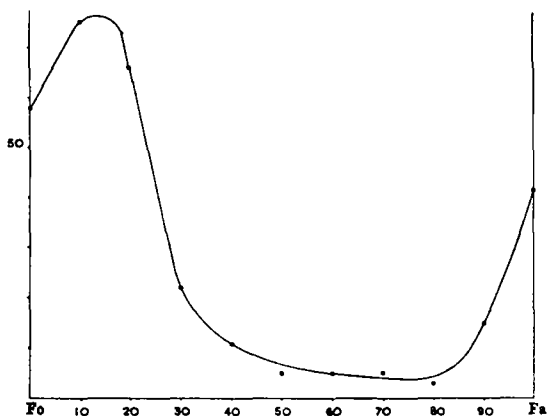


FIG. 4. Frequency-distribution curve for 257 analyses of olivines recalculated into weight percentages of forsterite and fayalite.

and fayalite. Neither the results of the work of Bowen and Schairer (37) nor the data provided in the present paper tend to support this suggestion. In the zoned olivine from the Rapa luscladite (table II, nos. 75–85) the composition runs from chrysolite to hortonolite (20Fa–70Fa) and from hortonolite to fayalite (58Fa–96Fa). Olivine from the Fair Head dolerite-pegmatite (table II, nos. 46–48) also partly covers the supposed immiscibility gap. The apparent scarcity of olivines in the interval between Mg-hortonolite and fayalite may be due to the difficulty of procuring suitable specimens.

There have been several attempts to correlate the composition of olivines with the composition of the containing rocks. B. Doss (4), who believed that olivines tend to develop a magnesium-rich outer zone, explained this phenomenon as being due to an early crystallization of iron-ore and a consequent enrichment of the residual magma in magnesia. On the other hand, M. Stark (23) arrived at the conclusion, which is now shared by many petrologists, that the residual magma is progressively enriched in FeO at the expense of MgO, and that the olivines show a parallel tendency. Stark has demonstrated that olivines in limburgites are richer in magnesia than olivines in basalts, and that olivines

in acid rocks are very rich in iron (23, p. 38). F. Rinkel (25) has also concluded that olivines from peridotites are the richest in magnesia and those from eulysite are the richest in iron, while those from basalts and gabbros occupy an intermediate position. The origin of eulysite is still a debatable point, but according to the original interpretation by H. von Eckermann (22) eulysite represents a pegmatitic residual solution of basic magma, i.e. a late crystallization product. In this connexion it is interesting to inquire into the nature and composition of fayalite-bearing rocks. L. Hawkes (24) has fully discussed this subject and published a table of 10 analyses of these rocks. To these I have added 12 recent analyses of fayalite-bearing granite, granophyre, porphyry, and phonolitic trachyte. The average and the range of composition of these 22 analyses gave:

| | | | | | | |
|---|-----|-----|-----|-----|--------|---------------|
| SiO ₂ | ... | ... | ... | ... | 65.20% | (56.59-75.55) |
| Na ₂ O | ... | ... | ... | ... | 5.42 | (3.21- 7.59) |
| K ₂ O | ... | ... | ... | ... | 4.58 | (1.59- 6.63) |
| FeO(Fe ₂ O ₃):FeO(Fe ₂ O ₃)+MgO = 0.95:1. | | | | | | |

This shows that fayalite-bearing rocks are alkalic and range from intermediate to acid, and, in the majority of cases, are quartz-bearing. There are, however, certain basic rocks in which olivine and quartz are co-present (olivine-bearing quartz-dolerites), but olivine in these rocks, as a rule, is only moderately rich in iron, although in some varieties, according to V. Sobolev (32), it may contain as much as 80Fa. The average of three analyses of olivine-bearing quartz-dolerite gives: SiO₂ 52.10, Na₂O 2.54, K₂O 1.19%, Fe/Mg ratio 0.73; and in this respect they do not differ from normal quartz-dolerites. According to Bowen and Schairer, fayalite can exist in equilibrium both with quartz and albite, as is actually the case in fayalite-bearing rocks; but in the olivine-bearing quartz-dolerites, olivine is too magnesian to be in equilibrium with quartz and its presence is explained better if we assume that it represents an untransformed relict of an early phase of crystallization. Therefore, one may agree with Bowen and Schairer when they say that the fayalite-bearing rocks 'are always rich . . . in alkali feldspars alone or together with quartz or nepheline and thus belong among the trachytes, rhyolites, and phonolites' (43, p. 408). Recently, A. B. Edwards (44, p. 299), from his measurements of the optic axial angle of olivines from a suite of rocks ranging from limburgite to trachyte, also comes to the conclusion that olivine becomes more fayalitic as the magma approaches the trachytic end of the suite.

To find the relation between the composition of the olivine and the

composition of the parent rock I have calculated the Fe/Mg ratio for some of the rocks studied in this paper and the corresponding ratios of olivines as derived from their optical data. The results of these calculations are given in table IV and fig. 5.

TABLE IV. Calculated ratio $\text{FeO}(\text{Fe}_2\text{O}_3) : \text{FeO}(\text{Fe}_2\text{O}_3) + \text{MgO}$ for rocks and olivines.

| | No. | Rock-type. | Rock. | Olivine. |
|--|---|----------------------------------|-------|----------|
| Irish Tertiary rocks. | 1. Glomeroporphyritic aggregates (= allivalite, Rum), Fair Head | ... | 0.27 | 0.37 |
| | | 2. Olivine-dolerite, Fair Head | 0.56 | 0.57 |
| | | 3. Dolerite-pegmatite, Fair Head | 0.65 | 0.82 |
| | | 4. Olivine-dolerite | 0.57 | 0.38 |
| | | 5. Olivine-basalt | 0.57 | 0.42 |
| British Carboni- ferous- Permian rocks | 6. Teschenite | ... | 0.64 | 0.40 |
| | | 7. Olivine-dolerite | 0.60 | 0.49 |
| | | 8. Essexite | 0.57 | 0.56 |
| | | 9. Luscladite, Rapa | 0.46 | 0.76 |
| | 10. Fayalite-bearing rocks | ... | 0.95 | 0.98 |

The data for the Rapa luscladite have been obtained from the analysis published by W. Campbell Smith and L. J. Chubb (26), that for the Fair Head dolerite and dolerite-pegmatite from unpublished analyses by the author. Unfortunately, there is no analysis as yet of the glomeroporphyritic aggregates from the Fair Head sill and so an analysis of allivalite from the Isle of Rum was taken instead, as the rocks are very similar in composition. The remaining ratios were derived from the calculated averages of the appropriate analyses.

The diagram (fig. 5) provides us with a clear picture of the relation between olivines and rocks in respect of their relative proportions of FeO and MgO. In the majority of dolerites and basalts the olivines are richer in MgO than their parent rocks. Considering that a fairly large proportion of iron is removed from the magma in the form of ore this is what one would expect. Also from the nature of the thermal equilibrium diagram one would expect the early formed olivine to be richer in magnesia. The three points corresponding to the three phases of the Fair Head sill clearly show this effect. They also show the progressive enrichment in iron of the olivines in the successive cooling phases, corresponding to the enrichment in iron (relative to magnesia) in the rock as a whole.

T. F. W. Barth (30, p. 380) has given a similar diagram in which the amount of olivine present in the rocks is also shown. His conclusions

are that in the olivine-rich rocks olivine is more magnesian than in the olivine-poor rocks, and that the olivines of the phenocrysts are always richer in magnesia than those of the groundmass. The material given in the present paper is in complete agreement with the conclusions of Barth.

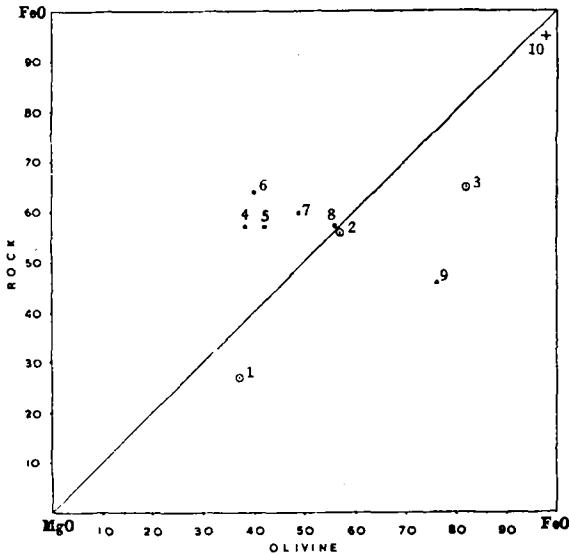


FIG. 5. Diagram showing the relation between the ratio $\text{FeO}(\text{Fe}_2\text{O}_3) : \text{FeO}(\text{Fe}_2\text{O}_3) + \text{MgO}$ calculated for the olivines and their parent rocks.

- | | | |
|---|--|-------------------------|
| 1. Glomeroporphyritic aggregates (= allivalite) | } Fair | } Irish Tertiary rocks. |
| 2. Olivine-dolerite | | |
| 3. Dolerite-pegmatite | } sill | |
| 4. Olivine-dolerite | | |
| 5. Olivine-basalt | | |
| 6. Teschenite | } British Carboniferous-Permian rocks. | |
| 7. Olivine-dolerite | | |
| 8. Essexite | | |
| 9. Luscladite, Rapa, Austral Islands. | | |
| 10. Fayalite-bearing rocks. | | |

In this respect olivines are similar to pyroxenes, the composition of which, according to A. Weich (16), also depends on the composition of the rock—in basic rocks pyroxenes are relatively rich in magnesia and get progressively richer in iron as the acidity of the rock increases.

All this shows that there exists an intimate connexion between the composition of the magma and the composition of olivines crystallizing

out of it at different stages of solidification or differentiation. There is also a parallelism between the compositional change in individual crystals of olivine as shown by zoning and that of the successive crops of olivine crystals in a cooling magma. Without inquiring into the actual process of differentiation of magma, which may be different in different cases, one must stress the importance of the control exercised by the magma on the minerals and the reciprocal control exercised by the minerals on the magma.

In conclusion, I wish to thank Lt.-Col. W. Campbell Smith, Dr. C. A. Anderson, Dr. A. D. N. Bain, Dr. C. E. Marshall, and Dr. A. C. Waters for sending me rock-specimens.

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