# Geochemical indicator of the efficiency of fractionation of the Skaergaard intrusion, east Greenland

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SUMMARY. When magmatic fractionation involves the settling and removal of crystals from the body of magma, the efficiency of the fractionation process may be defined as the degree of separation of the solid from the liquid phase. An expression is given that relates efficiency to the amount of mesostasis, or crystallized trapped liquid, in an igneous cumulate. The uranium contents of samples from a 349-m-long drill-core of part of the lower and hidden zones of the Skaergaard intrusion are used as a quantitative indicator of the amounts of mesostasis in the cumulates. There are marked changes in the amount of mesostasis over the length of the core and the average efficiency of fractionation was 85 %.

CRYSTAL settling is recognized as an important process in the formation of many layered and other igneous rocks (Wager and Brown, 1968), and is often considered to operate as the crystallization analogue of Rayleigh distillation (Rayleigh, 1896) in that the crystals are effectively removed from the melt as soon as they have formed. This concept has been used by a number of workers in predicting the variations of element concentrations in melt and solid fractions during fractionation, and has enabled conclusions to be reached on the petrogenesis of rock suites (Gast, 1968) or igneous complexes (Irvine and Smith, 1967). The unavailability of appropriate partition coefficient data for most elements has often necessitated the adoption of the assumption that element partition between the melt and a particular mineral phase remains constant during fractional crystallization. Information is often lacking also on the extent of the separation of the liquid and solid phases from each other, i.e. the efficiency of the fractionation process.

Our inability to predict how much pore material (i.e. crystallized trapped liquid) a cumulate contains stems not so much from lack of knowledge about crystal packing but about the extent of other processes such as adcumulus growth (Wager, Brown, and Wadsworth, 1960) or postdepositional re-equilibration. An assessment of the amount of pore material or mesostasis in any particular rock can be made from textural and mineralogical evidence (e.g. Wager, 1960) or, in certain cases, from appropriate geochemical data (e.g. Henderson, 1970). The present evidence (Wager and Brown, 1968; Henderson, Mackinnon, and Gale, 1971) suggests that there may be considerable variations in the amount of mesostasis, and hence of fractionation efficiency, from one layer to the next and from one layered intrusion to another. This paper presents an attempt to quantify the efficiency of fractionation during the forma-

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tion of part of the Skaergaard Layered Intrusion, East Greenland (Wager and Brown, 1968).

Fractionation efficiency (E) may be defined by the expression:

$$E_{\rm v} = 100(V_{\rm r} - V_{\rm m})/V_{\rm r}$$
 %,

where  $V_r$  = volume of rock, and  $V_m$  = volume of mesostasis (i.e. crystallized trapped liquid) for the case where efficiency is expressed in volumetric terms ( $E_v$ ). In so far as geochemical data are usually given in mass units, it might be preferable to express fractionation efficiency as:

$$E_{\rm m} = 100(\rho_{\rm r}V_{\rm r}-\rho_{\rm m}V_{\rm m})/\rho_{\rm r}V_{\rm r}\,\%$$

where  $\rho_r$  and  $\rho_m$  are the densities of the rock and mesostasis respectively. It is virtually impossible to determine the density of the mesostasis by direct means, and the value of  $\rho_m$  will not in many cases differ from that of  $\rho_r$ . It will, therefore, be assumed for the purposes of this paper that the approximation  $E_v \approx E_m$  holds.

The effect of fractionation efficiency on the rate of change in concentration of an element in a melt undergoing Rayleigh crystallization can be given by:

$$C_1/C_2 = F^{[0.01E_{\rm m}(k-1)]}$$

where: F = mass fraction of liquid remaining after fractionation,  $C_1 = \text{concentration}$ of the element in F,  $C_0 = \text{initial concentration}$  of element in melt, k = partitioncoefficient for the element, M, defined as  $[M]_{\text{cumulate phases}}/[M]_{\text{magma}}$ , and  $E_m = \text{fractiona$ tion efficiency, for the case when <math>k is a constant throughout the fractionation process. Greenland (1970) has discussed the effect on fractionation of variations in the partition coefficients of individual elements and in the modal proportions of the cumulus minerals. The equations derived in Greenland's paper can be readily modified to take account of fractionation efficiency.

The effect that different fractionation efficiencies would have on the chemistry of residual melts can be illustrated for a hypothetical case in which olivine is the only cumulus phase. Consider nickel and ferrous iron with the partition coefficients between olivine and magma taken to be 16 and 2.2 respectively (Henderson and Dale, 1969). Table I lists  $C_1/C_0$  values for different efficiencies and for different fractionation stages. Table II gives the ratio  $(C_1/C_0)_{\rm Ni}/(C_1/C_0)_{\rm Fe}$  for the different stages and shows the important role that efficiency can play in determining element ratios in residual melts (e.g. for the specific case where 60 % of the magma has fractionated (F = 0.4) the nickel to ferrous iron concentration ratio in the residual magma is over 40 times larger when fractionation efficiency has been 70 % than when it has been 100 %).

Samples and method. During the 1966 expedition to the Skaergaard intrusion, four drill cores were obtained from part of the layered series and adjacent country rocks. One of the drill sites was positioned on the northern bank of Uttentals Sund, below and to the south of Uttentals Plateau (fig. 1). The drilling at this site yielded 349 m of core ( $3\cdot 2$  cm diam.) comprised of rock from the hidden zone and lower zone of the intrusion. Miss Yin Yin Nwe of Cambridge University has estimated (private communication) that the bottom of the core is of material somewhere between 150 to

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E F	100 %		90 %		80 %		70 %	
	Ni	Fe	Ni	Fe	Ni	Fe	Ni	Fe
0.9	0.31	0.81	0.24	0.89	0.28	0.90	0.33	0.92
0.8	0.035	0.77	0.049	0.79	0.069	0.81	0.096	0.83
0.2	4·7×10 <sup>-3</sup>	0.65	8·1×10-3	0.68	0.014	0.71	0.024	0.74
0.6	4·7×10-4	0.24	$1.1  imes 10^{-8}$	0.28	$2.2  imes 10^{-3}$	0.61	4·6×10-3	0.65
0.2	3·1 × 10 <sup>-5</sup>	0.44	8.6×10-5	0.47	2·4×10-4	0.21	6·9×10-4	0.56
o·4	$1.1  imes 10_{-6}$	0.33	4.2 imes 10 <sup>-6</sup>	0.37	$1.7 \times 10^{-5}$	0.42	6.6×10 <sup>-5</sup>	0.46
0.3	$1.4 \times 10^{-8}$	0.24	8-7×10-8	0.27	5·3×10-7	0.32	3·2×10 <sup>-6</sup>	0.36
0.5	3·3×10 <sup>-11</sup>	0.14	3.7×10-10	0.18	4·1 × 10-9	0.21	4.6×10-8	0.26
0·1	1.0×10-12	0.06	3·2×10-14	0.08	1·0×10 <sup>-12</sup>	0.11	3.2×10-11	0.13

TABLE I.  $C_1/C_0$  ratios for nickel and ferrous iron at different fractionation stages and for different efficiencies (E)

TABLE II. Ratios of  $(C_l/C_o)_{Nl}/(C_l/C_o)_{Fe}$  at different fractionation stages and for different efficiencies (E)

Ε	100 %	90 %	80 %	70 %
$\overline{F}$		· · · · · · · · · · · · · · · · · · ·		
0.9	0.23	0.22	0.31	0.36
o·8	0.046	0.063	0.085	0.15
0.2	$7.2  imes 10^{-3}$	0.015	0.019	0.032
0.6	$8.7 \times 10^{-4}$	$1.8  imes 10^{-3}$	3.6×10-3	7·1×10-3
0.2	$7 \cdot 1  imes 10^{-5}$	1·8×10-4	$4.7 \times 10^{-4}$	1·2×10-3
o·4	3·3×10-6	I·I $ imes$ IO-5	$4 \cdot 1 \times 10^{-5}$	1·4×10 <sup>-4</sup>
0.3	5·9×10-8	3·2×10-7	$1.7  imes 10^{-6}$	8·9×10-6
0.5	$2.3  imes 10^{-10}$	2·I × 10 <sup>-9</sup>	$1.9 \times 10^{-8}$	1.8×10-7
0.1	1.6×10-14	$3.8  imes 10^{-13}$	9·I × 10 <sup>-12</sup>	7·3×10 <sup>-11</sup>

180 m below the base of the exposed Layered Series (Wager and Brown, 1968). Her estimate is based upon the compositions of the cumulus crystals in the drill core.

The samples taken from the drill core are plagioclase, olivine orthocumulates and mesocumulates with intercumulus clinopyroxene, iron ore, and apatite. The plagioclase crystals are commonly zoned in a fashion that is characteristic of those in the lower zone orthocumulates (see fig. 17a, Wager and Brown, 1968). The olivine crystals are generally small and often rounded. In all the specimens studied here plagioclase is the dominant cumulus mineral and,



FIG. I. Location of drill site on Skaergaard Intrusion. Map is based on Plate XI in Wager and Brown (1968).

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except for a few specimens, the modal proportions of the minerals: plagioclase, olivine, and pyroxene, do not vary greatly. The petrography and mineralogy of the drill core are currently under study at the University of Cambridge.

Wager and Deer (1939) observed that phosphorus did not enter appreciably into the composition of any of the cumulus minerals of the lower layered rocks of the Skaergaard intrusion and they believed that most of this element exists in the interstitial material or mesostasis. This led to the development of the use of certain elements present in cumulates as indicators of the relative amount of mesostasis (Wager, 1963; Wager and Brown, 1968; Henderson, 1968 and 1970). Henderson, Mackinnon, and Gale (1971) showed that uranium can be used in this way provided uraniumbearing cumulus minerals such as apatite are absent. This condition holds for the samples from the drill core as apatite is present only as an intercumulus mineral and has crystallized from the trapped liquid. The mesostasis, therefore, contains virtually all the uranium in these cumulates with negligible contribution from the cumulus minerals. If, as postulated by Wager, Brown, and Wadsworth (1960), the mesostasis of an igneous cumulate has the same composition as the contemporary magma and if this composition, or at least the uranium content, is known for various fractionation stages, then the proportion of mesostasis and the fractionation efficiency can be quantitatively determined.

Five centimetre lengths of half-core were taken at approximately 7.5-m intervals and, after removal of a slice for a thin section, were crushed to pass a 120 mesh. Between 2 and 3 g of powder were accurately weighed, in duplicate, into polythene vials, which were then sent, with standards, to A.W.R.E. at Aldermaston for uranium analysis by the delayed-neutron technique as described by Gale (1967). Gamma-flux from the short-lived radioisotopes (principally aluminium) present in the samples after irradiation can cause interference in the uranium analysis if the intensity reaches a certain level. Although it is possible to discriminate against this interference, the system used at A.W.R.E. for this work had not been designed to do this and it was necessary to keep the sample weights below the level which would cause gamma-ray interference. Unfortunately the low sample weight had an adverse effect on the delayedneutron counting statistics and increased the relative standard deviation of the results to about 20 %. This precision, although satisfactory for the work reported here, is not as good as can be obtained by this method (see Gale, 1967, for details).

Results. The average uranium concentrations of the samples are given in Table III.

In the case of the Skaergaard intrusion the data are available that make it possible to relate quantitatively the uranium content of a rock to the fractionation efficiency. Henderson (1970) has shown that the uranium concentration of the contemporary magma that gave rise to rocks at 100 m above the base of the exposed layered series was  $1 \cdot 1$  ppm. This result may be used to estimate the change in uranium content of the magma during the fractionation period represented by the core. For this to be done it is necessary to know the mass fractions of the Skaergaard magma that had crystallized by the beginning and by the end of the drill-core sequence. This, in turn, requires a knowledge of the size of the hidden zone, about which there is debate (Wager, 1960; Wager and Brown, 1968; Chayes, 1970). Chayes (1970) has computed three estimates of the magnitude (by volume) of the hidden zone based upon different schemes for weighting the zones of the exposed layered series. Of the three he shows a preference for the 81 % value but goes on to say (page 9): 'Indeed, the critical point is just that on the basis of presently available data no firm decision between the available alternatives is possible.' Current geophysical work may help to resolve this problem but in the meantime the figure of 81 % is adopted. Fortunately, as the figure of  $1 \cdot 1$  ppm. uranium in the magma at the 110-m position was obtained by a method that is independent of estimates of the size of the hidden zone, the final conclusions reached in this paper will not be significantly altered whatever reasonable estimate (including zero) is made of the proportion of the intrusion that is hidden.

TABLE III. Uranium contents of samples from Skaergaard drill-core

Depth	U ppm.	Depth	U ppm.	Depth	U ppm.	Depth	U ppm.	Depth	U ppm
2.9 m.	0.12	69·6 m.	0.16	137·2 m.	0.12	213.7 m.	0.18	281.8 m.	0.16
10.4	0.20	77.1	0.25	144.7	0.20	220.8	0.22	289.8	0.15
17.1	0.16	83.6	0.14	152.4	0.14	228.7	0.12	297.2	0.15
25.6	0.10	91.5	0.10	160.5	0.10	236.5	0.13	304.9	0.15
33.2	0-20	99.1	0.12	167.7	0.21	243.7	0.18	312.4	0.15
41.4	0.17	106.8	0.10	175.1	0.15	251.3	0.15	320.1	0.50
47.7	0.20	114.1	0.12	182.8	0.13	259.1	0.30	327.6	0.52
53.9	0.18	122.3	0.14	108.1	0.15	266.7	0.12	335.2	0.16
60 4	0.29	129.8	0.14	205.6	0.02	274.3	0.10	342.6	0.31

It needs to be noted that neither Chayes (1970) nor Wager (1960) was able to take into account the different densities of the rocks that make up the layered series, even though the geochemical data are expressed in mass units. For this paper it is taken that the hidden zone comprises 81 % by mass of the entire intrusion.

On the basis of Chayes's calculation it is estimated that the drill reached rock at about the 79.9 % (F = 0.2014) fractionated level and started at about the 82.5 % (F = 0.1747) level. In order to calculate the concentration change of uranium in the contemporary magma during the fractionation of the material of the core the equation (15a) given by Greenland (1970) is used:  $C_1/C_0 = F^{(p.k-1)}$ , where p is a proportionality constant and in this case is the proportion of trapped liquid in the fractionated rock sequence. This equation may be written in the more general form:

$$\ln(C_{12}/C_{11}) = (p.k-1)\ln(F_2/F_1).$$

As discussed by Greenland, the trapped liquid may be mathematically considered as another solid phase with k = 1. The uranium data in Table III, together with the value of 1·1 ppm. U in the contemporary magma at 110 m above the base of LZ<sub>a</sub>, indicates that the average amount of mesostasis in the rocks is about 15 % (i.e p = 0.15). The substitution of the appropriate data into the above equation gives:  $\ln(1.1/C_{1 \text{ base}}) = (0.15-1)\ln(0.1816/0.2014)$  for the base of the core to the 110-m position (F = 0.1816) within LZ<sub>a</sub>, from which  $C_{1 \text{ base}} = 1.007$  ppm. This is the uranium concentration of the magma that gave the earliest rock of the core. For the whole of the core:  $\ln(C_{1 \text{ top}}/1.007) = (0.15-1)\ln(0.1747/0.2014)$ , from which  $C_{1 \text{ top}} = 1.136$  ppm. U. Hence, during the fractionation of the magma to give the rocks of the core, the uranium concentration rose by about 0.13 ppm. This variation has been taken into account in calculating the percentage of mesostasis in the samples from the core



FIG. 2. Variation in percentage of mesostasis with depth in drill-core.

and the results are given in fig. 2. The average amount of mesostasis is  $15\cdot4$  % which is in line with the value assigned to p in order to solve the equations and, therefore, no iterations of the calculations are required.

Discussion and conclusions. Wager and Deer (1939, pp. 38-45) describe the nature of the rhythmic layering in the Skaergaard intrusion and report on its variation between the central part of the layered series and the marginal belt. Rocks in the central part are more leucocratic and have better-developed rhythmic layering than those near the

margins. The drill was positioned about 800 m from the western contact and was, therefore, off the cross-bedded belt at the margin of the layered series. However, the rocks from the core studied here are uniform and rarely show evidence of good rhythmic layering. Despite this, there are significant variations in the amount of mesostasis (between 5 and 26 %) over the length of the core with, in places, sudden and large changes (e.g. between 200 and 230 m depth. The sample taken from about 206 m depth, with about 5 % mesostasis, is slightly more melanocratic and has smaller laths of plagioclase than the other samples). Henderson, Mackinnon, and Gale (1971, Table 1) have recorded marked changes in the amount of mesostasis over short stratigraphical distances in the lower zone.

The average proportion of mesostasis is about 15% and, therefore, the fractionation efficiency over this stage was about 85%. This result confirms the earlier work (Wager, 1963), which showed that the lower zone rocks have more mesostasis than the middle and upper zones. However, the average value for the amount of mesostasis in this part of the intrusion, which includes some of the hidden zone, indicates that Wager's (1960) estimate of the hidden zone rocks containing about 35% mesostasis may well be too high. Evidence from a simple experiment (Wager and Brown, 1971, page 66) led Wager to believe that the initial packing of cumulus plagioclase crystals would result in a crystal mush containing at the most 60% of crystals and about 40% of intercumulus liquid. Adcumulus growth, or other processes such as filter press action, could expel some or virtually all of the liquid. If Wager's estimate of the initial amount of liquid is correct then some has been expelled during the formation of rocks of the core.

It does not appear possible to relate the changes in fractionation efficiency to causes other than effectively random variations in the nature of crystal packing and the subsequent loss of intercumulus liquid by adcumulus growth. Hence, it is impossible

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to make a meaningful prediction of the amount of mesostasis in any one rock. An average value for the fractionation efficiency during the genesis of various stages of a layered intrusion may, however, be helpful in elucidating some of the petrogenetic processes involved. The efficiency should be taken into account when estimates of the volume or mass proportions of fractionation stages of an intrusion are calculated (e.g. Chayes, 1970) or used to help interpret the geochemical behaviour of elements (e.g. Dissanayake and Vincent, 1972) and is a particularly important parameter when studying the geochemical distribution of the 'incompatible elements' as they will be concentrated in the phases of the mesostasis.

Far more quantitative work on fractionation efficiency in other layered intrusions is required in order to assess the role that it plays in the petrogenesis of layered igneous rocks.

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