# Cryptorhythmic and macrorhythmic layering in aegirine lujavrite, Ilímaussaq alkaline intrusion, South Greenland\*

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Cryptorhythmic layering (Wilson 1992) has been established in a 200 m drill core through a layered aegirine-eudialyte nepheline syenite, aegirine lujavrite I, of the Ilímaussaq alkaline intrusion. Within each cryptorhythmic unit, the content of Y in cumulus eudialyte is cons-tant, but there is a sharp increase to a new fixed value in the next unit. Each cryptor-hythmic unit contains from two to four macrorhythmic units which show moderate to poor density sorting and consist of three layers defined by successively high contents of cumulus aegirine, then cumulus eudialyte, microcline and nepheline, and finally intercumulus zeolites and arfvedsonite. The "dry" density of both cumulus and intercumulus material decreases in the higher cryptorhythmic units and is thought to reflect the decreasing density of the parental liquid layers. The appearance of a new cryptorhythmic unit does not coincide with the end of a macrorhythmic unit and is apparently unrelated to pulses of nucleation and settling out of cumulus phases. Crystallization of cryptorhythmic units from chemically distinct double-diffusive liquid layers in the aegirine lujavrite I magma and of macrorhythmic units by repeated in situ crystallization on the floor of the Ilímaussaq magma chamber is discussed.

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# Introduction

Igneous petrologists have long considered that fractional crystallization combined with the settling out of the fractionating phases was the dominant process in the differentiation of magmas (Bowen 1928; Wager 1963). The process can be demonstrated on the mgto-g scale in the laboratory (Bowen 1915) and in nearsurface lava lakes (Helz 1987) but its recognition in the geological record is largely a matter of interpretation rather than unequivocal demonstration. Indeed, the settling out of crystals in magma chambers has recently been challenged and there is a body of opinion which considers it unlikely (Campbell 1978; McBirney & Noyes 1979; Sparks et al. 1984).

In the present article, the process of fractional crystallization is monitored through 200 m of a macrorhythmically layered nepheline syenite (aegirine lujavrite I) from the Ilímaussaq intrusion, South Greenland. The lujavrite crystallized from a large syenitic pluton at a late stage in its evolution when the volume of the residual magma chamber was small and when the residual magma had reached an alkali-rich agpaitic, volatilerich state. It is demonstrated that Y and U can be used

Bailey: Cryptorhythmic and macrorhythmic layering

to reveal systematic shifts in the chemistry of cumulus eudialyte but these shifts do not coincide with the boundaries of macrorhythmic units.

# Geological setting

The Ilímaussaq intrusion (Fig. 1) forms part of a continental rift-zone province, the late Precambrian Gardar igneous province of South Greenland. Events began with a marginal intrusion of augite syenite and two thin sheets of peralkaline granite beneath the roof. The third, and most voluminous, pulse of magma was peralkaline and silica-undersaturated. It evolved to form a layered series of agpaitic nepheline syenites in the centre of the intrusion (Ussing 1912; Ferguson 1964; Bailey et al. 1981a; Larsen & Sørensen 1987).

The roof rocks of the third pulse - pulaskite, foyaite, sodalite foyaite and naujaite - crystallized successively downwards while contemporaneous bottom cumulates are thought to be present at depth. The exposed bottom cumulates, the kakortokites, contain blocks of the roof cumulates and are therefore younger. The kakortokites



Fig.1. Geological map of the Ilímaussaq intrusion after Ferguson (1964). The location of drill core 7 in aegirine lujavrite I is marked more accurately on Fig. 3.



Fig.2. A. Schematic section through the Ilímaussaq intrusion. B. The lowermost 800 m of the intrusion showing the detailed igneous stratigraphy of the kakortokites and lujavrites. C. ZrO<sub>2</sub> profile for drill core 7 in aegirine lujavrite I. High ZrO<sub>2</sub> contents correspond to eudialyte-rich cumulate layers. After Andersen et al. (1981a) and Bailey et al. (1981b).



Fig. 3. Outcrop map of aegirine lujavrite I in the southeastern part of the Ilímaussaq intrusion (Fig. 1) from Henriksen (1993). Only the more conspicuous eudialyte-rich layers can be followed in the field and more have been recognised by geochemical and modal logging of drill core 7.



Fig. 4. Modal (vol.%) and  $K_2O$  (wt.%) analyses, and macrorhythmic and cryptorhythmic layers, for drill core 7, aegirine lujavrite I, llímaussaq intrusion. Aeg - aegirine, eud - eudialyte, m+n microcline and nepheline, z+a zeolites and arfvedsonite; F inferred fault. Proposed boundaries and internal layering of 10 macrorhythmic layers also incorporate data on  $ZrO_2$  contents (Fig. 2). "Dry" densities of macro- and cryptorhythmic layers are based on modal analyses and measured mineral densities. Density symbols with arrows refer to incomplete layers where the overall density is expected to be higher than the measured. Note that the boundaries between cryptorhythmic layers as defined by the Y and U contents of eudialyte only coincide in one instance, at 152 m depth.

range through well layered, weakly layered and transitional layered varieties before passing conformably upwards into lujavrites which have been divided into several varieties (Fig. 2B). The transition between the kakortokites and lujavrites probably reflects physical changes in a shrinking magma chamber due to subsidence and fracturing of the roof, causing lujavrites to evolve independently in two or more sub-chambers and to locally intrude and brecciate the overlying naujaite roof.

The earliest of the lujavrite varieties is an aegirinerich nepheline syenite orthocumulate termed aegirine lujavrite I (Andersen et al. 1981a; Bohse & Andersen 1981; Bailey & Gwozdz 1994). It conformably overlies the transitional layered kakortokite in the southeastern part of the intrusion and extends for 4 km along the Lakseelv valley at the head of Kangerluarsuk fjord. It forms a sheet-like body thinning out towards the south where the naujaite roof extends to lower levels. The sheet ranges from about 50-300 m in thickness and averages around 80 m.

Drill core 7 (GGU number 108107) is located 1.0 km NE of the mouth of Lakseelv at an altitude of 120 m and was obtained from the southern end of aegirine lujavrite I near its middle levels (Figs. 1, 2C, 3). It did not penetrate the underlying kakortokites. Xenoliths of the overlying naujaite roof occupy about 10% of the core. Since the lujavrite dips at  $45 +/-10^{\circ}$  both in the surface outcrops and within most of the core (assumed to be vertical), the 200 m of core probably represents a true stratigraphic thickness close to 140 m. However, the uppermost 18 m of the core is a faulted repetition of deeper levels. The present dip of the core can be related to tilting associated with nearby faults, and the slope of the original depositional floor is unknown.

# Analytical methods

Drill core 7 was divided into 1 m intervals which were crushed in an agate mortar for 30 minutes. Zr and Y were determined by X-ray fluorescence spectrometry (XRFS) on powder pellets using the techniques of Norrish & Chappell (1977). Interference from Sr K $\alpha$ and Rb K $\beta$  on Zr K $\alpha$  and Y K $\alpha$ , respectively, was corrected for. Matrix variation was monitored by measuring the Mo K $\alpha$  Compton peak in an off-peak position (-0.15 °2 $\theta$  using a LiF (220) crystal) to avoid interferences from Nb and Y lines.

Precision for Zr and Y was inside 1% relative and results were standardised against USGS reference materials (G-2, GSP-1, AGV-1, W-1 and BCR-1) using the recommended values of Govindaraju (1994). Determined (and recommended) values in ppm for the international lujavrite standard NIM-L are Zr 10600 (11000), Y 20.1 (22).

U was analysed by delayed neutron analysis at the Risø National Laboratory. Imprecision typically ran-

ges from +/-3% at the 10 ppm level to +/-2% at the 40 ppm level.

The remaining elements in this report were determined by instrumental neutron activation (REE, Cs, Th, Hf, Ta, Co, Sc), a combined neutron activation and Cerenkov counting technique (Li, F), and a variety of XRFS methods.

Mineral separation was effected by a combination of heavy liquid and magnetic techniques. Impurities (0.5-4 vol.%) were visually estimated and analyses adjusted accordingly.

Modal analyses are based on counting about 500 points in one thin section per metre. Bulk "dry" densities of rocks are based on modal analyses and on mineral densities which were determined on calibrated mixtures of heavy liquids.

## Macrorhythmic layering

Macrorhythmic layering (Irvine 1987) in aegirine lujavrite I was first recognised in drill core 7 on the basis of Zr (eudialyte) contents coupled with hand specimen and microscope observations (Bailey et al. 1981b; Bailey & Gwozdz 1994) and was later confirmed by detailed mapping (Henriksen 1993). It is now reappraised using modal analyses and contents of  $K_2O$  as a monitor of microcline abundances.

Unlike the spectacular macrorhythmic layering of the lower layered kakortokites (Ussing 1912; Sørensen 1969; Ferguson 1970; Bohse et al. 1971), aegirine lujavrite I only exhibits moderately to poorly densitystratified macrorhythmic units. These consist of three individual layers but, unlike the kakortokites, they are successively rich in cumulus aegirine, then cumulus eudialyte, microcline and nepheline, and finally intercumulus material (zeolites and arfvedsonite) (Fig. 4).

Allowing for the dip in drill core 7, individual macrorhythmic units have real thicknesses of 6-17 m; their three constituent layers grade into each other and are typically 3-8 m thick. The boundary to the next macrorhythmic unit is always gradual, commonly over several decimetres.

All three individual layers show planar lamination due to alignment of the platy microcline and acicular aegirine in the plane of layering. Although the uppermost layer crystallised from material rich in intercumulus liquid, it is still a cumulate rock in terms of its fabric and orthocumulate texture. Signs of current action such as scoured out troughs and current bedding have not been observed, and cumulus crystals are not linearly aligned.

The macrorhythmic units have been labelled M0 to M10 (Fig. 4). Unit M0, at the bottom of drill core 7, is incompletely represented. Unit M1 includes a very eudialyte-rich layer whose red colour can be easily followed in the field. Samples from 0-18 m depth, on the basis of layer thickness, modal contents and trends,

Table 1.	. Whole	e-rock and	d mineral	separate	analyses,	aegirine	lujavrite	I, sample	146 m.
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	Whole-rock	Aegirine	Eudialyte	$M + N^1$	Arfvedsonite	$A + N^2$
SiO	51.69	52.43	50.56	63.97	50.64	47.09
TiO	0.25	0.43	0.0	0.03	0.43	0.03
ZrO	1.48	0.24	13.63	0.05	0.06	0.06
Nb,Ó,	0.15	0.02	1.12	0.01	0.02	0.02
RE <sub>2</sub> O <sub>2</sub>	0.59	0.05	4.85	0.03	0.03	0.03
Al,Ô, °	10.92	0.28	0.23	18.38	0.43	25.97
Fe <sub>2</sub> O <sub>2</sub>	12.05	30.04	_	0.15	13.02	0.33
FeO	2.08	1.35	2.38 <sup>3</sup>	0.06	20.67	0.02
MnO	0.25	0.07	1.64	0.05	1.02	0.10
MgO	0.01	0.02	0.02	0.01	0.03	< 0.01
CaO	1.35	0.96	8.82	0.04	0.34	0.24
Na <sub>2</sub> O	11.01	12.67	12.72	0.65	8.13	15.62
K,Ô	2.80	0.01	1.00	15.85	2.56	0.66
Lí <sub>o</sub> O	0.032	0.007	0.002	0.001	0.429	0.007
P,Ô,	0.02	< 0.01	. –	< 0.01	< 0.01	<0.01
H,O	0.31	0.414	_	0.294	1.794	9.164
H,O⁺	3.70	-		-	-	_
F	0.06	< 0.05	0.05	< 0.05	0.28	0.11
Sum	98.73	99.09	97.07	99.54	99.90	99.41
Zr(ppm)	10800	2500	100900	130	570	160
Y	964	19	5750	31	8	15
U	23.4	6.2	143	3.5	3.7	5.0

Analysts: I. Sørensen, H. Bohse, J.C. Bailey, R. Gwozdz.

1 microcline + nepheline; 2 analcime + natrolite; 3 total Fe as FeO; 4 LOI.

All elements by  $\hat{X}RFS$  except Li and F by INA-CC, U by DN, FeO by the metavanadate technique, Na<sub>2</sub>O by atomic absorption, and H<sub>2</sub>O and LOI by gravimetry.

bulk density and the Y-U chemistry of eudialyte (see below), are interpreted to represent a faulted repetition of unit M7. Although the upper boundary of the underlying M10 unit has been faulted, M10 seems to be virtually complete.

Since several eudialyte-rich layers can be traced over an area of at least  $1.3 \times 0.3 \text{ km}$  (Fig. 3) (Henriksen 1993), it seems likely that the macrorhythmic units established in core 7 have a considerable lateral extent.

The decision to locate the start of a macrorhythmic unit at the onset of an aegirine layer carries with it the important consequence that each macrorhythmic unit shows a rough density sorting with an aegirine layer (s.g. around 2.80 g.cm<sup>3</sup>) followed by a eudialyte and microcline + nepheline layer (c. 2.75) and a layer rich in intercumulus material (c. 2.73). The range of density values is smaller than the range for the black (c. 3.07), red (c. 2.79) and white (c. 2.77) layers of the kakortokites (Ferguson 1970). Furthermore, in aegirine lujavrite I, the eudialyte layers show no clear signs of being located below the microcline + nepheline layers (Fig. 4). Thus density sorting can only be characterized as moderate to poor.

Bulk densities of the 10 complete macrorhythmic units show an overall decrease towards the higher units (r = 0.59) (Fig. 4). This is due to (a) the falling propor-

tion of cumulus phases in the upper units (r = 0.62) especially the two densest - aegirine and eudialyte (r = 0.67), (b) the decreasing density of the bulk cumulus phases (r = 0.21) and (c) the decreasing density of the intercumulus material (r = 0.37) due to increasing amounts of zeolites (r = -0.65) towards the higher units. Post-magmatic zeolitization partly explains some of these features but the same trends have also been noted in the least zeolitized samples (Bailey & Gwozdz 1994).

## Petrography

Aegirine lujavrite I shows a marked textural and mineralogical distinction between the cumulus phases (aegirine, eudialyte, microcline, nepheline) and the intercumulus material (arfvedsonite, natrolite, analcime) (Bailey & Gwozdz 1994). Aegirine needles, microcline plates and the nearly equant eudialyte and nepheline crystals are all euhedral to subhedral; they are set in a matrix of interstitial to subpoikilitic natrolite and analcime together with ovoids (oikocrysts) of arfvedsonite. Primary igneous minerals and textures are commonly replaced by the matrix zeolites. The most

Sample Rock	D7 146 m aeg luj I	D7 96 m aeg luj I	D7 46 m aeg luj I	D7 4 m aeg luj I	D7 33.7 m naujaite	
SiO	50.56	49.86	49.06	50.13	49.01	
TiO	0.05	0.05	0.03	0.06	0.07	
ZrO	13.63	13.34	13.79	13.03	13.29	
HfO,	0.18	0.16	0.17	0.17	0.23	
Nb,Ô,	1.12	1.14	1.24	1.08	1.09	
Ta,Õ,	0.04	0.04	0.04	0.04	0.06	
RĚ <sub>2</sub> O <sub>3</sub>	4.12	4.42	4.87	4.10	1.86	
Al <sub>2</sub> O <sub>3</sub>	0.23	0.34	0.48	0.41	0.68	
FeO*	2.38	3.05	3.16	3.74	6.08	
MnO	1.64	1.88	1.77	1.65	1.07	
MgO	0.02	0.03	0.07	0.02	0.03	
CaO	8.82	8.37	8.03	8.37	10.48	
Na <sub>2</sub> O	12.72	13.19	12.66	13.00	13.24	1. A.
K,Ô	1.00	0.65	1.02	0.61	0.25	
F	0.05	< 0.05	0.12	0.09	<0.05	
Cl	0.94	0.87	0.83	0.87	1.33	
Sum	97.50	97.39	97.34	97.37	98.77	
Li(ppm)	8.5	7.5	13	32	17	
Rb	281	116	147	172	39	
Sr	695	749	702	694	476	
Pb	778	197	666	871	246	
Ba	905	968	790	1100	913	
La	6210	6880	7540	6090	3010	
Ce	14600	16300	17600	14500	6560	ίę. κ
Pr	1930	2170	2280	1930	806	
Nd	6440	7240	7660	6460	2620	· .
Sm	1330	1510	1530	1340	506	
Gd	1370	1530	1550	1390	586	
Dy	1300	1430	1410	1300	644	
Ho	282	296	286	272	156	
Er	756	826	796	758	434	
Yb	704	759	734	698	417	
Ľu	100	106	101	94	61	
Y, ·	5750	6410	6410	5770	2810	
Th .	65	85	74	88	35	
<b>U</b>	143	174	156	154	52	
Zn	699	93	993	1060	130	÷.,-
La/Yb <sub>N</sub>	5.82	6.00	6.78	5.76	4.77	
Th/U	0.45	0.49	0.47	0.57	0.67	

Table 2. Analyses of eudialytes from aegirine lujavrite I and an enclosed naujaite xenolith, Ilímaussaq intrusion.

common accessory phases are acmite, albite, sphalerite, native lead and iron oxides, whereas Li-mica (polylithionite) and molybdenite are only occasionally visible. Naujaite xenoliths, sometimes intensely altered, have contaminated adjacent lujavrites in a variety of ways. Patches and veins of zeolites together with shear, fracture and breccia zones commonly with later powdery coatings can be assigned to post-magmatic stages.

Sub- to euhedral eudialyte crystals, around 1 mm in diameter, are present at a background level of 2-5 vol.% in the core but some 11 eudialyte-rich layers containing maxima of 10-35 vol.% eudialyte also occur. The eudialyte crystals may include a few percent of aegirine needles up to 0.1 mm in length, that is about 10 times smaller than the cumulus aegirine crystals of the rock. Most eudialytes are optically homogeneous but occasionally one or two oscillatory zones may be present towards the outer margin. More rarely it is possible to observe patchy zoning of cores, non-concentric zoning, a single marginal zone or weak sector zoning. Overgrowths, amounting to about 5% of an individual crystal, may develop at one of the sides or corners of a eudialyte crystal but are also uncommon. In zeolitised samples, eudialyte grains are commonly strained or weakly to intensely cracked with slight dispersal of the fragments, and show alteration patches of natrolite,

Analysts: H. Bohse, J.C. Bailey, R. Gwozdz. All elements were determined by XRFS except for Li and F by INA-CC. Low sums are due to non-determination of H,O.

Table 3. Analyses of coexisting minerals in two samples of aegirine lujavrite I.

Sample	96 m	191 m								
Mineral	aeg	aeg	eud	eud	m+n	m+n	arf	arf	a+n	a+n
Cs	<0.3	<0.8	<3	<3	2.6	1.8	<0.9	1.7	37	 42
Rb	4	3	47	_	3110	2570	79	104	114	128
K%	0.87	0.11	0.35	0.38	13.2	12.5	2.15	2.24	0.41	0.48
Li	16	10	5.0	8.6	8.2	5.9	2090	2020	21	32
Ca%	0.68	0.92	6.18	7.34	0.01	0.04	0.24	0.28	0.16	0.28
Sr	6	6	777	654	10	11	17	14	8	10
Pb	96	275	180	990	260	160	40	600	23	718
La	52.6	32.3	6590	6530	20	30	12.2	24.7	37.3	26.7
Ce	118	74.4	16300	15600	47	63	33.7	55.7	81.6	65.2
Nd	_	_	7500	7030	-	-	_	_	-	_
Sm	9.1	6.2	1540	1560	2.2	2.9	1.9	3.6	5.2	4.8
Eu	0.58	0.47	141	129	0.11	0.15	_	-	0.25	0.36
Tb.	_	-	218	219	0.52	0.55	-		0.59	0.68
Yb	5.35	3.31	686	707	1.6	1.9	3.16	4.77	1.67	1.16
Lu	0.90	0.78	103	110	0.25	0.29	0.68	0.85	0.22	0.14
Y	41	28	6700	7140	20	22	29	29	23	16
Th	6.9	14.6	100	127	9.4	8.4	0.54	4.0	3.1	11.6
U	5.6	3.9	174	130	10.4	2.6	2.8	3.9	2.6	3.2
Zr%	0.21	0.25	9.87	10.21	0.01	0.01	0.07	0.07	0.01	0.02
Hf	55.3	62.6	1410	1760	5.3	3.1	18.4	21.4	2.7	5.6
Ta	2.0	1.0	370	420	1.7	0.5	1.5	1.2	1.2	0.8
Mg%	0.04	0.02	0.02	0.03	0.01	< 0.01	0.04	0.04	0.02	<0.01
Co	<0.3	<0.2	<1.0	<0.9	<0.2	<2.9	1.6	1.2	<3.0	<0.1
Zn	172	385	31	102	43	119	553	540	57	250
Sc	0.3	0.3	<0.1	<0.1	<0.1	<0.1	<0.1	0.4	<0.1	<0.1
Ga	56	54	<1	<1	76	70	33	32	61	46
K/Rb	220	370	75	-	42	49	270	215	36	31
La/Yb <sub>N</sub>	6.5	6.4	6.3	6.1	8.3	10.4	2.5	3.4	14.7	15.2
Th/U	1.2	3.7	0.6	1.0	0.9	3.2	0.2	1.0	1.2	3.6
Zr/Hf	38	39	70	58	24	38	39	31	41	- 43

Analysts: R. Gwozdz, H. Kunzendorf, J.C. Bailey.

Minerals: aeg - aegirine; eud - eudialyte; m+n - microcline + nepheline; arf - arfvedsonite; a+n - analcime + natrolite. All elements were determined by INA except for Rb, K, Ca, Sr, Pb, Y, Zr, Mg, Zn and Ga by XRFS, Li by INA-CC and U by DN.

analcime, a Na-Zr-REE phase (?catapleiite) and zirfesite.

## Mineral chemistry

Analyses of rock-forming minerals in aegirine lujavrite I are characterized by low-temperature end-member compositions (Table 1). Thus MgO contents in aegirine and arfvedsonite are at trace-element levels, microcline is virtually Na<sub>2</sub>O-free and eudialyte is the end-member of the eucolite-mesodialyte-eudialyte series (Vlasov 1966). In the absence of major element substitutions, normal methods of checking cryptic variations in an igneous sequence using electron microprobe techniques are unsuitable.

However, cryptic variations are present among trace elements, notably REE, Th, U, Hf and Nb in separated eudialytes (Table 2). Contents of these elements in eudialyte increase in the higher layers of aegirine lujavrite, though there is a reversal in the sample from the highest level at 4 m depth in the core where faulting has repeated the igneous stratigraphy (see below).

Additional trace element analyses on separated minerals are presented in Table 3 and indicate that the bulk of REE, Th, U, Zr, Hf, Nb and Ta is located in eudialyte. Preliminary work thus suggests that cryptic changes can be followed by analysing these elements in eudialyte but that the changes are only around a few percent relative and would be difficult to monitor by microprobe or neutron activation analytical techniques. XRFS analyses have the required precision but it was found that mineral separates, even of high purity, are not fully representative of a mineral phase because of selective losses during the separation procedures, e.g. crushing and removal of rims and alteration areas from eudialytes. However, at the n00-n000 ppm levels found for several of these elements in these modally variable rocks, it is possible to perform whole-rock analyses with high precision (see Analytical Methods) and to infer mineral chemistry with a high degree of confidence.

## Whole-rock analyses of Zr, Y and U

Contents of Zr, Y and U in one metre samples from drill core 7 through aegirine lujavrite I are plotted in Fig. 5. There is a clear coherence between the three elements due to their marked concentration in eudialyte and thus in eudialyte-rich layers. There are no obvious, overall trends for Zr and Y through the drill core but U maxima and minima tend to increase upwards from 30 to 40 ppm and 12 to 15 ppm, respectively.

Eudialyte-rich layers have higher Y/Zr but lower U/ Zr ratios than other layers and this causes these ratios to vary with the abundance of eudialyte (not shown). Overall, these ratios increase in their maximum and minimum values through the drill core suggesting that the higher eudialytes are richer in Y and U.

Plots of Zr-Y and Zr-U (Fig. 6) indicate that a number of linear arrays are present. Each array is restricted to a specific depth interval in the drill core and the various arrays are interpreted as step-like shifts in the chemistry of eudialyte. Each line unites samples with fixed contents of Y and U in eudialyte, and the lines can be extrapolated to the Zr content of eudialyte (10.0 wt.%) in order to calculate these contents (Table 4).

The inferred Y contents (9900 to 11550 ppm) and U contents (118 to 210 ppm) of eudialytes increase at higher stratigraphic levels. For both elements, contents obtained by extrapolation generally exceed those found by analysis of separated eudialytes (Table 2). This feature was noted by Bailey et al. (1983) and Bailey & Gwozdz (1994), and can be attributed to loss of Y and U during mineral separation when the easily crushed, altered grains of eudialyte and the U- (and probably) Y-rich rims of eudialytes tend to be lost. It is important to emphasise, however, that within whole-rock samples such Y and U must have been retained in situ throughout crystallization and (in many samples) during postmagmatic zeolitization; only by such retention can one explain the highly regular Zr-Y and Zr-U arrays of Fig. 6.

En masse, the remaining rock-forming minerals also show step-like shifts in their Y and U contents. At the Zr content of eudialyte-free lujavrite (2500 ppm), and according to the Zr-Y arrays, the Y contents of eudialyte-free lujavrite increase in a series of steps towards the higher stratigraphic levels of the core (Table 4). The same conclusion is reached for U though the innate scatter of the Zr-U arrays adds an air of uncertainty.

The percentage standard deviation of Zr and Y contents about each Zr-Y regression line ranges from about 1% at high Zr-Y levels to 2% at lower levels

and can thus be explained by the combined imprecision of the Zr and Y analyses (see Analytical Methods).

Only four samples out of 179 lie more than 36 away from their inferred Zr-Y regression lines and these deviations are mainly explained by post-magmatic alteration. Zr-U relations are less regular. In order to establish the Zr-U regression lines of Fig. 6 it was necessary to reject 32 out of 179 samples. This procedure was justified by (a) the reasonable degree of Zr-U coherence displayed by the remaining 147 samples and (b) the clear linkage found between the deviant samples and the occurrence of post-magmatic zeolitization. For example, zeolitized samples in the sequence from 125.5–129.5 m have all lost some U.

Only the implications of the regular Zr-Y and Zr-U relations are considered further.

The levels of step-like shifts for Y and U do not correspond to the junctions between macrorhythmic units. Instead, the Y steps established at 174, 152, 122 and 88 m depth all lie in eudialyte-rich layers, being located in the lower, middle, middle and upper parts of these layers, respectively. The smooth rise and fall of Zr and the modal trends in these layers show no sign of any deviation at the step-like boundaries. Furthermore, most eudialyte layers are not associated with these boundaries.

In only a single example does the position of an Y step coincide with the position of a U step. This is at 152 m depth where the Y step is reliably established to 152 + i/- 0.5 m and the U step is less securely established to 152 + i/- 2 m.

Steps in the Y and U contents of eudialyte are all positive. The only apparent reversal is in the uppermost 18 m of the drill core. Here, Y contents of eudialytes are indistinguishable from those at 89.5–121.5 m depth (Fig. 6) and U contents can be matched with those from 18.5-105 m depth. Thus, the uppermost 18 m of the drill core corresponds to stratigraphic levels between 89.5 and 105 m, a conclusion already reached on the basis of modal variations and attributed to repetition by faulting (see above). Although two faults parallel to the lujavrite layering have been recognised just south of drill core 7 (Fig. 3), smaller faults are difficult to recognise in these fissile lujavrites and, at the moment, field control is inadequate to confirm or deny the inferred stratigraphic repetition.

The conclusion that Y and U contents in eudialyte can be repeated by faulting, and can be integrated with a similar faulted repetition of the macrorhythmic layering, suggests that the steps in Y and U contents are also stratigraphic features and define the boundaries of stratified compositional units. On the basis of steps in the Y contents of eudialyte, these units have been labelled C0 to C4 (Fig. 4).

The steps in Y contents of eudialyte are generally around 4% relative, and are roughly eight times less pronounced than those for U. There is a weak tendency (r = 0.34) for the Y steps to be greater at higher levels in the core and a similar tendency seems to occur for

Cryptorhythmic unit	C3	C4	C3	C2	C1	C0
Depth (m)	0.5–17.5	18.5-87.5	89.5-121.5	122.5–151.5	152.5-173.5	174.5-197.5
Real thickness (m)	>13	48	22	20	15	>16
Y in eudialyte (ppm) <sup>1</sup>	11100	11550	11100	10700	10200	9900
Y in eudialyte (ppm) <sup>2</sup>	7790	8000 8200	7690	7380	-	7140
Y at 2500 ppm Zr <sup>1</sup>	107	152	107	88	81	72
Depth (m)	0.5-17.5	18.5-105	105-150	151.5–164	180-197.5	
U in eudialyte (ppm) <sup>1</sup>	210	210	152	115 <sup>3</sup>	118	
U in eudialyte (ppm) <sup>2</sup>	154	152, 156 174	143	-	130	
U at 2500 ppm Zr <sup>1</sup>	11.4	11.4	10.7	11.23	8.4	

Table 4. Cryptorhythmic units as defined by contents of Y and U in cumulus eudialyte, aegirine lujavrite I, Ilímaussaq intrusion.

See Fig. 4 and text for discussion.

1 - by extrapolation from Zr-Y and Zr-U arrays (Fig. 6); 2 - XRFS (Y) and DN (U) analysis of individual separated eudialytes; 3 - unreliable values because of limited points on Zr-U array.

the U steps also. At the moment, however, the number of steps available for checking this trend is barely adequate.

The sizes of the chemical steps appear to be correlated with the thicknesses of the stratigraphic units they define. Thus, the lowest unit for Y where bottom and top are both defined is only 15 m thick, but the overlying units are successively 20, 22 and at least 48 m thick. The top of this last unit is thought to be terminated by a fault and may originally have been greater. Against this apparent regularity, however, one should note that below the lowest defined unit there is an incomplete unit which is at least 16 m thick. The thicknesses of U units are less reliably defined but also seem to increase towards the higher stratigraphic levels of the core.

Setting on one side the incomplete Y unit C0, it is found that from unit C1 to C4 there is a steady decrease (r = 0.88) in the bulk "dry" density from 2.828 to 2.727 g.cm<sup>3</sup> (Fig. 4). Since the density of both cumulus (r =0.94) and intercumulus phases (r = 0.34) in units C1 to C4 decreases, it seems reasonable to infer that the densities of the parental magmas for the Y units were also decreasing.

#### Discussion

#### Cryptorhythmic layering

Wilson (1992) informally proposed the term "cryptorhythmic layering" to describe the presence of discrete geochemical arrays over specific stratigraphic intervals in cumulus bronzitites of the layered Great Dyke, Zimbabwe. Within a single array, an incompatible element such as Ti increases with differentiation of the bronzite, i.e. with falling Mg/Mg + Fe, reflecting its uptake during the reaction between adcumulus bronzite and trapped intercumulus liquid. Other arrays show the same trend but at different absolute contents of Ti, and thus point to compositional discontinuities in the magma chamber. It was suggested that the series of cryptorhythmic units developed by crystallization from a series of double-diffusive convection layers in the Great Dyke magma chamber.

The step-like changes in the chemistry of eudialyte and, en masse, in the other rock-forming minerals in aegirine lujavrite I satisfy the criteria for cryptorhythmic layering and are also thought to reflect crystallization from a series of compositionally distinct liquid layers.

A more rigorous linkage between the chemistry of eudialytes and coexisting magmatic liquids would require analysis of eudialyte cores only. In the technique used here, the inferred contents of Y in eudialyte are bulk contents for cores and rims as well as rare overgrowths and alteration patches. The geochemical regularity within an individual array apparently demands that both the overall crystallization history of eudialytes within the array and the composition of the coexisting magmatic liquid were constant.

The simplest explanation for the step-like shift to the next array is that there was also a step-like shift in magma composition. A more complex alternative is to argue that all the lujavrite magmas and coexisting eudialyte cores represented by drill core 7 were constant in composition but that the subsequent crystallization history of eudialyte rims etc. had a step-like character. This would imply that amounts and/or compositions of intercumulus and/or post-magmatic liquids shifted in a step-like manner. Petrographic evidence of such steps, however, has not been found.



Fig. 5. Zr, Y and U contents in drill core 7, aegirine lujavrite I. Gaps in the sequence are occupied by xenoliths from the naujaite roof rocks.

Furthermore, step-like changes in the amount of intercumulus magma and of final residues of this magma (Bailey & Gwozdz 1994) show no relation to the cryptorhythmic boundaries established here.

Unlike the bronzites of the Great Dyke (Wilson 1992), the levels of Y in the lujavritic eudialytes are constant within a single cryptorhythmic unit. In part, this is thought to reflect the orthocumulate character of aegirine lujavrite I and the lack of interaction between the cumulus eudialyte and the intercumulus liquid, but it also points to the absence of fractionation processes within a cryptorhythmic unit.

The thicknesses of the best defined cryptorhythmic units in the Great Dyke range from 3–30 m. Units in aegirine lujavrite I tend to be thicker, ranging from 15–>48 m for Y and from roughly 9–>61 m for U.

The presence of liquid layers at the aegirine lujavrite I stage of the Ilímaussaq magma chamber receives support from the decreasing bulk "dry" density of the rocks in the higher cryptorhythmic units. It is agreed that the only stable arrangement for liquid layers in a magma chamber is one of decreasing density at higher levels and that density differences of <1% (at least in simple aqueous systems) can be maintained by double-diffusive convection (Turner and Gustafson 1981). Differences of 0.5–2.4% rel. are observed in the "dry" rock density of adjacent layers in drill core 7.

The decrease in aegirine contents -21.4 to 14.6 vol.% equivalent to a decrease from 28.3 to 19.9 wt.% and a density decrease of 0.302 g.cm<sup>-3</sup> - more than explains the decrease in rock densities of the cryptorhythmic units from 2.828 to 2.727 g.cm<sup>-3</sup>. The sharp density decrease reflecting the fall in aegirine abundance is mainly compensated by the rising proportion of intercumulus material, especially the zeolites. These changes in mineral proportions point to decreasing levels of Fe and increasing H<sub>2</sub>O in successive liquid layers.

Macrorhythmic layering

One model of fractionation in magma chambers (Wager and Brown 1968) considers that dense cumulus phases nucleate near the cold roof of a chamber, and then sink or are carried by convection currents to the chamber floor. On this model, density-sorted macrorhythmic units represent the arrival of a pulse of crystals and should be integrated with cryptic changes in the cumulus phases.

In aegirine lujavrite I, the onset of macrorhythmic units and shifts in the chemistry of cumulus eudialyte are not integrated. Conceivably, a single pulse of crystallization could settle out as several macrorhythmic units reflecting the periodic arrival of a single crystal-charged current, but even here the step-like shift should occur at the end of the last deposited unit rather than in the middle of a unit as observed.

The origin of macrorhythmic units in the lower

layered kakortokites at Ilímaussaq was considered by Larsen and Sørensen (1987) and the reader is referred to their discussion. Largely by analogy with recent ideas on liquid layering in magma chambers, they proposed that one or several complete macrorhythmic units fractionated from an individual double-diffusive liquid layer until density equalization occurred with the overlying liquid layer. If similar processes occurred in aegirine lujavrite I, then density equalization may have been delayed as a result of the lower bulk density of these cumulates relative to the kakortokites. Furthermore, because the bulk density of the aegirine lujavrite I macro- and cryptorhythmic units in drill core 7 decreased with time, an increasingly delayed equalization of density would be expected and might explain the tendency for more macrorhythmic units to occur in the upper cryptorhythmic units of the drill core.

In the case of aegirine lujavrite I, the model of Larsen and Sørensen (1987) fails to explain why fractional crystallization within a macrorhythmic unit did not lead to shifts in the composition of cumulus eudialyte in the succeeding macrorhythmic unit. Contents of Y in cumulus eudialyte are apparently constant even in a sequence of four macrorhythmic units within cryptorhythmic unit C4.

As an alternative, solidification of a liquid layer may have proceded not by fractional crystallization but by an extreme case of in situ crystallization during which residual magma escaped in such small amounts from the crystallizing cumulates that the composition of the liquid layer did not detectably evolve (Langmuir 1989).

In situ crystallization seems intuitively compatible with several features of the macrorhythmic units in aegirine lujavrite I: the subdued modal variations, the moderate to poor density sorting and the absence of a sharp onset to each macrorhythmic layer. It could proceed by a process of oscillatory nucleation (Maaløe 1978; Hort et al. 1993), though McBirney and Noyes (1979) argue that such nucleation occurs over scales of a few cm up to one m rather than over thicknesses of 6-17 m as found in macrorhythmic layers in agairing lujavrite I. Furthermore, the crystallization is not truly oscillatory since, after crystallization of the eudialyte, microcline and nepheline layer, there is development of a cumulate layer relatively rich in intercumulus material rather than an immediate return to aegirine crystallization. An extreme, non-fractionating variety of in situ crystallization, of course, would fail to generate a residual, less dense magma and another mechanism must be appealed to in order to generate the succession of apparently lighter macrorhythmic units in aegirine lujavrite I.

It is conceivable that in situ crystallization can be reconciled with the formation of macrorhythmic units by varying nucleation and growth rates. As Parsons and Becker (1987) emphasise for the Klokken intrusion, physicochemical controls on nucleation and growth rates are poorly understood and no attempt is made here to extend their calculations to the more



Fig. 6. Zr-Y and Zr-U compositional arrays for whole-rock samples in core 7, aegirine lujavrite I. Individual arrays are restricted to specific depth intervals and were defined by least-squares fitting; they represent cryptorhythmic layers which are considered to indicate the presence of liquid layers in the Ilímaussaq magma chamber.

esoteric magmas and minerals at Ilímaussaq. One should note, however, that this type of mechanism approaches an in situ character when (a) crystallization takes place in a magma chamber or layer of small vertical dimension thus minimising both collective and differential crystal settling and (b) the minerals involved are relatively small and have only minor density differences from the magma. The presence of liquid layers in aegirine lujavrite I, the smaller size of the lujavrite cumulus crystals compared to both the underlying kakortokites and the Klokken syenites, and the relatively low density of eudialyte (2.88 g cm<sup>-3</sup>) all suggest that eudialyte crystallized more or less in situ. In agreement with this, eudialyte-rich layers do not underlie and cannot be separated from, as in the kakortokites, the layers rich in crystals of microcline

+ nepheline (s.g. 2.55 g.cm<sup>-3</sup>). Nevertheless, in situ crystallization does not explain the moderate density grading within macrorhythmic units and must be modified to allow for some settling of crystals under gravity.

## Development of cryptorhythmic layering

The step-like geochemical shifts between compositionally distinct liquid layers may have been set up by sidewall crystallization leading to the rise of lighter, fractionated liquids towards the roof zone of the magma chamber (cf. McBirney 1980; Turner and Campbell 1986). This process would have been minimised at the kakortokitic stage of the Ilímaussaq intrusion when the intrusion had a large aspect ratio of 8-50 (Bailey et

Bailey: Cryptorhythmic and macrorhythmic layering

al. 1981a) but would have accelerated at the lujavritic stage when, after roof foundering, the Ilímaussaq chamber rapidly converted to two or more sub-chambers with lower aspect ratios. Chemically, these liquids should be enriched in Y and U relative to Zr since at this stage in the Ilímaussaq chamber Y and U have bulk partition coefficients <1 whereas Zr has a coefficient >1 (Andersen et al. 1981b). Furthermore, because fractionation takes place at an exceptionally evolved stage of the Ilímaussaq system (probably >95% crystallization) and in a sub-chamber of restricted volume, the rising sidewall liquids will show a large increase in their contents of incompatible elements (including Y and U) and these increases will be transferred to the liquid layers and, on crystallization, to their cumulus eudialyte.

Most of the preceding discussion has assumed that the cryptorhythmic units in aegirine lujavrite I are best defined by shifts in the Y contents of eudialyte. Shifts in U contents only coincide with those for Y in one instance and seem to demand a different origin. Previous workers (e.g. Steenfelt & Bohse 1975, Andersen et al. 1981b) have appealed to sudden losses of volatiles and complex ions such as  $UF_6^{2-}$  to explain unusual features in the distribution of U in Ilímaussaq materials. Similar processes, operating at the aegirine lujavrite I stage, may have influenced the transfer of U in sidewall liquids or perhaps from one liquid layer to the next.

Finally, one is left with a series of questions on why cryptorhythmic units always terminate and start in eudialyte-rich layers. Although eudialyte is a less dense mineral than aegirine, its chemical components are around 24% heavier than those of again and it will thus be more effective in lowering the density of the host magma and triggering density equalization with the overlying liquid layer. This influence, however, is more than offset by the much greater abundance of cumulus aegirine. Is there a fixed delay between the period of aegirine-rich crystallization and the moment of density equalization in the succeeding eudialyterich layer? Does in situ oscillatory crystallization proceed across cryptorhythmic boundaries regardless of the changes in major- and trace-element chemistry inferred between one liquid layer and the next? And why are there no obvious modal or petrographic changes when crystallization starts in a new liquid layer? Clearly there is considerable scope for further research on fundamental aspects of layering in the Ilímaussag intrusion.

## Tentative conclusions

Cryptorhythmic layering has been identified by monitoring the chemistry of cumulus eudialyte in a drill core through the earliest lujavrite (orthocumulate aegirine lujavrite I) of the Ilímaussaq intrusion and is taken as evidence for the presence of double-diffusive liquid layers. The appearance of cryptorhythmic layering is linked to foundering of the intrusion's roof, the appearance of a sub-chamber with a sloping roof, a lower aspect ratio and a smaller vertical interval than the preceding kakortokitic stage when bottom cumulates probably formed over the full extent of the intrusion.

The double-diffusive liquid layers, with lower densities but higher Y/Zr and U/Zr ratios at higher stratigraphic levels, may have been established by ascent of a low density fractionated magma along the sloping roof. The extreme fractionation of such agpaitic magmas promoted detectable cryptic changes in the liquid layers and their cumulus eudialyte.

In contrast, the internal crystallization of each cryptorhythmic unit did not lead to internal cryptic changes in eudialyte chemistry suggesting that crystallization on or near the floor of the lujavrite sub-chamber occurred by an extreme in situ, rather than a fractional, process.

Crystallization of each cryptorhythmic unit took place via the formation of two to four macrorhythmic units but the boundaries of the two types of layering do not coincide.

Ten macrorhythmic units have been recognised in the lujavrite drill core. Each cumulate unit consists of three sub-layers enriched in aegirine, then in eudialyte, microcline and nepheline, and finally in intercumulus phases (zeolites and arfvedsonite). They are characterized, relative to the macrorhythmic units of the earlier lower layered kakortokites, by subdued modal variations, moderate-to-poor density sorting and a diffusely marked start. It appears that these features are largely consistent with in situ crystallization though the moderate to poor density sorting requires some settling of crystals under gravity.

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# Dansk sammendrag

Kryptorytmisk lagdeling (Wilson 1992) er påvist i en 200 m lang borekerne gennem en lagdelt ægirineudialyt nefelin syenit, ægirin lujavrit I, fra den alkaline Ilímaussaq intrusion. Indholdet af Y i kumulus eudialyt

er konstant inden for hver kryptorytmisk enhed, men ændres brat til en højere værdi ved overgangen til en ny enhed. Hver kryptorytmisk enhed indeholder fra to til fire makrorytmiske enheder, der viser moderat til dårlig sortering efter vægtfylde, og består af tre lag defineret ved successivt høje indhold af kumulus ægirin, fulgt af kumulus eudialyt, mikroklin og nefelin og endelig interkumulus zeoliter og arfvedsonit. Den "tørre" vægtfylde af både kumulus og interkumulus materialer aftager i de øvre kryptorhytmiske enheder, hvilket antages at afspeile aftagende vægtfylde "modervæskens". Fremkomsten af en ny kryptorytmisk enhed falder ikke sammen med afslutningen af en makrorytmisk enhed og har tilsyneladende ingen relation til nukleation og bundfældning af kumulus faser. Krystallisation af kryptorytmiske enheder fra kemisk veldefinerede dobbelt diffusive væskelag i magmaet for ægirin lujavrit I og af makrorytmiske enheder ved gentagen in situ krystallisation på Ilímaussaq magmakammerets bund diskuteres.

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Bailey: Cryptorhythmic and macrorhythmic layering

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