Supplementary information

Sample details

Samples used were from the Natural History Museum, London, UK: collections BM1968 P37 and BM1957 1056, and are listed in Supplementary Table1 and Table 2.

Supplementary Table 1

Chilwa Island fenite samples. Last three samples used only for mineralogical investigation, and do not have whole-rock analyses

NHM number	NHM rock type classification	Fenite grade used in this study
BM1968 P37 63	Quartz fenite	Medium
BM1968 P37 71	Quartz fenite	Low
BM1968 P37 72	Quartz fenite	Low
BM1968 P37 78	Quartz fenite	Medium
BM1968 P37 83	Quartz fenite	Not graded, probably high
BM1968 P37 96	Quartz fenite	Medium
BM1968 P37 101	Quartz fenite	Medium
BM1968 P37 130	Quartz fenite	Low
BM1968 P37 32	Syenite fenite	Medium
BM1968 P37 54	Syenite fenite	Medium/high
BM1968 P37 68	Syenite fenite	Medium/high
BM1968 P37 102	Syenite fenite	Medium/high
BM1968 P37 139	Breccia	Breccia
BM1968 P37 146	Breccia	Breccia
BM1968 P37 100	Quartz fenite	Medium
BM1968 P37 126	Quartz fenite	Medium
BM1968 P37 137	Syenite fenite	Medium/high

Supplementary Table 2

a) Chilwa Island carbonatite samples used in whole-rock analyses

NHM Number	NHM rock name
BM1957 1056 59	Sövite
BM1957 1056 73	Sövite
BM1957 1056 120	Pyrochlore sövite
BM1957 1056 90	Pyrochlore & ankerite in sövite
BM1968 P37 170	Dolomitic/ankerite carbonatite
BM1957 1056 96	Ankeritic carbonatite
BM1957 1056 94	Ankeritic carbonatite
BM1957 1056 122	Ankeritic carbonatite
BM1957 1056 114	Sideritic carbonatite
BM1968 P37 172	Sideritic & REE-rich carbonatite

b) Chilwa Island carbonatite samples used for mineralogical investigation

NHM Number	NHM rock name
BM1957 1056 102	Ankeritic carbonatite
BM1957 1056 118	Pyrochlore-rich carbonatite
BM1957 1056 128	Pyrochlore-rich carbonatite
BM1957 1056 113	Sideritic carbonatite

Methodology

Back-scattered electron imaging (BSE), cathodoluminescence (CL) imaging and quantitative and mineral composition analyses were carried out at Kingston University on a Zeiss EVO 50 scanning electron microscope (SEM) and Oxford Instruments analytical suite (INCA) comprising an INCA X-act spectrometer and a CL Gatan Chroma-CL imager. The EDS operating conditions were an accelerating voltage of 20kV with a beam current of 1-1.8 nA. Spot size is controlled by the beam current and was approximately 5 µm. Calibration was made to a Co standard. Standards used are set out in Supplementary Table 3. Analyses of RE-bearing minerals are set out in Supplementary Tables 4-7.

Supplementary Table 3 Standards used for SEM

Element	Standard
F	Barium fluoride
Na	Jadeite
Mg	Periclase
Al	Corundum
P	Apatite
Cl	KCl
K	Orthoclase
Ca	Wollastonite
Ti	Ti metal
Fe	Fe metal

Calibration standards for the REE are phosphate glasses, with a chemistry of (REE)P_4, tested against a series of Drake and Weill (1972) REE glasses analyses, which yielded robust results.

Wavelength dispersive electron microprobe analyses of apatite and RE minerals were carried out using a JEOL JXA-8200 instrument at the Camborne School of Mines, University of Exeter. Operating conditions were an accelerating voltage of 20kV, a current of 28 nA and a 1 µm diameter beam size. TAP, PET and LIF, with a variety of natural mineral standards were used for calibration together with a ZAF matrix for background positions. Each element was measured for 20s on the peak and 10s on the background positions. The Pr L-beta line was used to avoid overlap with La. Background positions for the REE were adjusted to avoid overlaps and empirical correction factors were used for overlapping peaks. Fluorapatite was used as the calibration standard for Ca and P to minimise matrix effects and to reduce problems of overlap between P and F peaks. Detection limits were calculated as 3 standard deviations above background. Small spot size (5 µm) and a relatively high current were used to obtain reasonable analyses on very small minerals. Over 30 analyses of F were made to compensate for any crystal orientation effect. Element mapping of apatite was done at 20 kV, 33 nA and a 1 µm beam size.

Spatially resolved trace element determinations of a variety of apatites were obtained by Laser Ablation Inductively Coupled Mass Spectrometry (LA-ICP-MS) on a New Wave UP193FX Laser Ablation System coupled to Agilent 7500cs ICP-MS at the Natural History Museum in London. NIST 612 was used as an external standard, and SiO_2 as an internal standard, with Si values determined by SEM/EDS. Data were collected for 90s, using spot sizes of 20-30 µm, and laser fluence of 3.2J/cm^2 firing at 10Hz. Analytical errors on this instrument are considered to be below 2-3%.

Whole rock analyses (Supplementary Table 8) were carried out at the Natural History Museum in London. Each sample was pre-treated with 1 ml concentrated HNO_3, and then fused with 120 mg of $LiBO_2$ in a Pt/Au crucible. Trace elements were measured by quadrupole inductively coupled mass spectrometry (ICP-MS). An aliquot of 100 mg of each sample was pre-treated with concentrated HNO_3 and dissolved in a mixture of 4 ml HF and 1 ml $HClO_4$ at 100˚C. This solution was dried down at 150˚C and the residue redissolved at 150˚C using 2 ml $HClO_4$. The solution was again dried down, and then redissolved in a mixture of 1 ml concentrated HNO_3 + 1 ml H_2O + 0.5 H_2O_2 at 70˚C, and afterwards made up to 10 ml with water. Nonisobaric interferences in the ICP-MS analyses were reduced by tuning CeO/Ce^+ to <1% and Ba^{2+}/Ba^+ to <1%. For Eu, Gd, Yb, Hf and W, the levels of polyatomic interferences were estimated using single-element standards and calculated concentrations were corrected accordingly. The concentration of Ga (using ^{71}Ga) was corrected for $^{142}REE^{++}$. Certified reference materials BCR-1, BHVO-1 (basalts, USGS standards), JG-1 (granodiorite, Geological Survey of Japan standard), JLS-1 (limestone, Geological Survey of Japan standard), GA (acid granite, CRPG standard), SY-3 (syenite, CCRMP standard), DNC-1 (dolerite, USGS standard) and MAG-1 (marine mud, USGS standard) were used to monitor the accuracy of all element analyses. Comparison with standards was made once per sampling run.
Loss on ignition (LOI) data was obtained at the Camborne School of Mines.

Raman spectroscopic analysis of double-polished wafer was carried out at Kingston University using a Renishaw RM1000 instrument with CCD detection via WIRE 1.3 software. A silicon standard with a single peak at 521 cm^{-1} was used, and runs were for 20s using a 514 nm Ar ion laser.

Microthermometry of fluid inclusions was performed at Kingston University, using a heating and cooling TMS600 mechanical stage attached to a Nikon Optiphot instrument, with a Tm94 Controller and Linksys 32 software. Calibration was made against synthetic fluid inclusion standards containing “pure” CO_2 and H_2O. The reported temperatures for the stage shoud be accurate to ± 2˚C for temperatures between 50˚C and 350˚C, ± 0.5˚C for temperatures between 0˚C and 50˚C, and ± 0.2˚C for temperatures between -60˚C and -1˚C.

Supplementary Table 4 SEM-EDS analyses of apatite in fenite and carbonatite

Low-grade n=3	SiO2	CaO	SrO	Na2O	P2O5	La2O3	Ce2O3	Pr2O3	Nd2O3	F	O = F	Total
Representative	0.77	52.87	1.47	0.61	38.87	0.48	1.19	n.d.	0.64	3.58	1.51	98.97
Std deviation	0.40	0.90	0.24	0.42	0.21	0.11	0.12		0.10	0.47		
Medium-grade rim n=12												
Representative	1.62	51.89	1.19	0.06	39.72	0.49	1.23	0.23	0.75	3.49	1.47	99.20
Std deviation	0.39	0.62	0.23	0.02	0.11	0.01	0.16	0.01	0.17	0.19		
Medium-grade core n=12												
Representative	0.83	53.29	0.67	0.07	41.39	0.16	0.42	0.05	0.29	3.75	1.57	99.35
Std deviation	0.27	0.30	0.27	0.03	0.28	0.04	0.12	0.02	0.11	0.18		
Medium/high grade RE-rich n=15												
Representative	1.04	50.17	2.53	0.75	39.73	1.10	2.00	0.16	0.57	3.42	1.44	100.03
Std deviation	0.31	0.22	0.92	0.11	0.64	0.37	0.39	0.08	0.12	0.43		
Medium/high grade RE-poor n=4												
Representative	n.d.	52.90	2.79	0.53	40.78	0.48	1.15	0.15	0.45	2.42	1.02	100.63
Std deviation		2.13	0.81	0.24	0.77	0.09	0.04	0.09	0.15	0.79		
Carbonatite n=7												
Representative	0.63	52.18	2.73	0.22	40.97	0.13	0.36	n.d.	0.10	3.13	1.32	99.13
Std deviation	0.11	0.86	0.39	0.13	0.42	0.09	0.14	n.d.	0.07	0.67		

Note: Y not analysed n.d. = not detected

Supplementary Table 5 SEM-EDX analyses of monazite-(Ce) in fenite and carbonatite

Low-grade n=10	SiO2	FeO	CaO	SrO	P2O5	Y2O3	La2O3	Ce2O3	Pr2O3	Nd2O3	Sm2O3	Gd2O3	Dy2O3	ThO2	F	O = F	Total
Representative	n.d.	1.11	1.29	0.66	28.72	1.28	15.79	33.05	3.21	12.14	1.32	1.20	n.d.	0.28	0.42	0.18	100.29
Std deviation		0.17	0.54	0.20	0.48	0.88	1.59	0.36	0.45	1.83	1.04	0.99		0.25			
Medium-grade n=25																	
Representative	0.72	0.96	1.21	0.54	28.55	n.a.	15.27	32.33	3.05	12.30	1.19	0.92	n.a.	2.12	n.d.		99.16
Std deviation	0.48	0.63	0.52	0.51	1.43		2.97	2.02	0.56	2.56	0.69	0.47		1.48			
Rock 83																	
Normal Ce	0.80	2.20	3.80	0.29	28.15	6.71	12.47	17.69	2.45	9.38	1.74	1.59	1.11	5.74	n.d.		94.12
Low Ce	0.87	1.33	2.97	0.16	27.90	8.96	17.54	3.55	3.16	13.80	2.80	1.96	3.61	3.53	n.d.		92.14
Carbonatite n=2																	
Representative	0.86	n.d.	1.51	3.43	28.46	n.a.	18.16	30.52	2.33	7.24	0.78	0.42	n.a.	4.91	0.13	0.05	98.70
Std deviation	0.85		0.60	0.06	0.23		0.15	0.81	0.20	0.12	0.32	0.40		0.26	0.12		

Note: CO_2 and HREE not analysed. HREE commonly ~2-3% in rock 83. 83 results are affected by mineral porosity and are considered to be semi-quantitative FeO = total iron n.d. = not detected n.a. = not analysed

Supplementary Table 6 SEM-EDX analyses of bastnäsite-(Ce) in fenite and carbonatite

Medium-grade n=7	**SiO2**	**FeO**	**CaO**	**SrO**	**Y2O3**	**La2O3**	**Ce2O3**	**Pr2O3**	**Nd2O3**	**Sm2O3**	**Gd2O3**	**Dy2O3**	**ThO2**	**F**	**O = F**	**Total**
Representative	n.d.	1.20	3.58	0.40	n.a.	19.74	28.30	2.58	9.03	1.48	1.06	0.60	1.79	10.84	4.56	76.04
Std deviation		0.76	0.57	0.35		2.92	1.25	0.70	2.26	1.07	0.57	0.44	0.72	1.54		
Rock 83 n=3																
Representative	1.80	1.04	3.02	0.49	2.70	10.21	34.30	2.05	6.04	1.23	0.79	0.94	9.99	7.25	3.05	78.80
Std deviation	1.06	0.95	0.70	0.18	0.56	6.26	17.77	1.03	3.48	0.56	0.72	0.16	2.19	0.56		
Ankeritic carbonatite n=6																
Representative	n.d.	0.44	3.53	1.17	n.a.	22.91	31.48	1.89	5.44	0.25	n.a.	n.a.	2.31	11.43	4.81	76.04
Std deviation		0.39	2.50	0.33		1.43	2.97	0.49	1.04	0.11			1.82	0.06		

Note: CO_2 & HREE not analysed n.d. = not detected n.a. = not analysed

Supplementary Table 7 SEM-EDX analyses of parisite-(Ce) in fenite and carbonatite

Medium-grade n=8	**SiO2**	**FeO**	**CaO**	**SrO**	**Y2O3**	**La2O3**	**Ce2O3**	**Pr2O3**	**Nd2O3**	**Sm2O3**	**Gd2O3**	**Dy2O3**	**ThO2**	**F**	**O = F**	**Total**
Representative	0.73	0.80	12.61	1.33	0.97	11.09	21.48	2.30	9.24	1.57	0.96	0.82	3.25	9.74	4.10	72.79
Std.deviation	0.44	0.54	3.73	0.36	0.86	4.74	2.65	0.67	2.66	0.59	0.44	0.79	1.63	2.43		
Ankeritic carbonatite n=2																
Representative	n.d.	0.63	10.03	0.87	n.a.	18.89	27.05	1.81	5.64	0.39	n.a.	n.a.	0.73	11.03	4.64	72.43
Std.deviation		0.59	1.47	0.14		3.51	0.85	0.30	0.93	0.10			0.27	1.95		

Note: CO_2 & HREE not analysed n.d. = not detected n.a. = not analysed

Supplementary Table 8 Whole rock analysis of fenite

BM1968 P37	72	130	71	32	63	78	96	101	54	68	102	139	146	83
Majors %														
SiO2	71.73	72.90	68.80	63.98	62.34	63.37	62.62	60.74	62.53	58.55	61.75	62.40	60.57	86.21
TiO2	0.08	0.12	0.24	1.21	1.16	1.03	1.18	1.15	0.90	1.02	0.80	0.09	0.04	0.07
Al2O3	15.87	13.53	14.39	13.49	13.75	13.96	14.57	13.13	14.14	12.61	13.56	16.55	16.44	0.49
Fe2O3 (t)	1.31	1.78	1.83	5.05	6.91	5.80	6.82	6.44	6.19	6.18	5.94	1.46	1.72	1.93
MnO	0.01	0.05	0.06	0.12	0.08	0.09	0.09	0.13	0.16	0.29	0.12	0.03	0.01	0.10
MgO	0.06	0.14	0.14	1.02	0.79	0.74	0.81	0.95	0.40	0.94	0.81	0.01	0.01	0.02
CaO	1.35	1.53	0.47	2.68	2.27	2.55	3.26	3.12	1.60	4.52	2.84	1.04	2.21	1.95
Na2O	4.25	6.13	3.35	5.19	3.88	4.09	4.46	4.62	5.77	5.18	5.29	0.22	0.27	0.05
K2O	5.75	2.14	7.58	5.30	4.88	5.19	5.15	4.86	6.00	6.30	5.50	14.08	13.60	0.05
P2O5	0.13	0.04	0.06	0.33	0.39	0.32	0.37	0.34	0.26	0.39	0.36	0.94	1.84	1.64
BaO	0.08	0.05	0.52	0.23	0.41	0.37	0.26	0.34	0.43	0.30	0.27	0.11	0.26	0.11
SrO	0.04	0.02	0.03	0.06	0.05	0.05	0.05	0.09	0.04	0.08	0.06	0.14	0.09	0.10
LOI	0.67	0.59	0.72	0.95	0.66	1.26	1.10	1.79	1.07	2.24	2.13	0.57	0.69	0.94
Total	101.34	99.024	98.19	99.61	97.571	98.82	100.74	97.70	99.50	98.59	99.44	97.64	97.77	93.66

F ppm	262	414	209	1167	523	1091	969	1303	1809	4181	2457	869	1521	2285
Cl ppm	250	177	187	167	292	182	217	187	247	203	205	114	116	n.d.

Traces ppm	72	130	71	32	63	78	96	101	54	68	102	139	146	83
Li	3.8	6.5	8.2	10.3	5.9	10.5	17.6	18.5	8.5	7.5	32.5	0.6	0.7	17.0
S	75.0	77.4	690.0	107.0	432.0	302.0	142.5	1184.6	644.0	197.0	255.0	294.0	139.0	749.0
V	23.4	6.9	41.3	27.6	49.0	35.4	56.3	97.6	99.8	216.8	51.7	46.8	47.5	72.2
Co	1.6	1.8	2.6	6.9	7.4	7.1	23.8	8.8	5.3	7.0	7.2	2.7	2.1	3.6
Cu	2.1	0.3	0.4	9.6	14.8	3.2	7.5	4.8	7.3	6.9	7.9	3.3	1.8	171.5
Zn	16.3	49.6	42.8	90.9	105.0	104.0	121.0	142.8	108.0	162.0	112.0	16.3	20.7	62.4
Ga	21.4	19.1	18.8	18.3	21.6	22.5	28.4	22.1	22.2	18.9	20.4	54.7	33.7	n.d.
Rb	132.4	31.0	187.3	105.4	81.2	87.0	85.3	86.0	93.5	99.3	86.2	198.9	224.1	1.1
Y	8.1	23.5	60.8	51.5	46.5	49.6	67.3	54.3	38.2	38.6	37.3	65.7	134.8	339.7
Zr	48.5	100.7	313.7	1000.0	1175.0	1238.0	52.5	1097.0	960.2	466.1	920.5	112.8	843.1	21.0
Nb	1.9	10.0	48.5	82.9	39.8	45.4	39.5	85.3	58.4	71.1	62.3	59.8	895.0	22.9
Mo	0.8	0.7	8.4	2.3	3.0	4.4	3.6	6.4	12.1	9.9	8.7	31.1	2.0	10.4
Hf	1.7	0.6	1.5	5.0	0.5	1.2	1.5	2.6	9.9	10.7	5.0	2.0	0.8	0.2
Ta	0.1	0.5	0.9	4.5	1.6	2.7	1.6	2.9	0.9	1.9	1.8	0.7	16.2	n.d.
W	0.6	0.5	1.0	2.3	3.8	1.8	155.8	2.4	3.4	1.9	6.7	1.1	6.9	0.4
Pb	24.5	10.8	29.1	34.3	18.9	17.6	14.0	23.1	85.2	39.7	25.1	14.9	6.0	263.2
Th	1.6	2.7	7.1	11.0	2.3	5.0	4.0	6.2	24.2	9.2	9.8	32.6	76.0	280.0
U	0.6	0.2	1.1	1.8	0.4	0.6	0.4	0.4	0.9	0.5	0.6	5.1	10.9	20.3

REE ppm	72	130	71	32	63	78	96	101	54	68	102	139	146	83
La	15.0	17.9	91.8	85.7	53.3	51.6	53.1	118.4	79.1	79.9	126.1	14.1	102.3	483.5
Ce	30.8	37.6	138.0	170.0	122.0	100.5	126.3	164.0	145.3	164.0	215.6	22.5	158.7	786.2
Pr	3.5	4.2	19.6	21.2	18.2	13.6	17.9	22.7	18.4	20.6	24.8	2.5	16.7	88.1
Nd	12.4	15.7	69.0	84.4	82.2	55.4	69.0	79.2	72.2	80.8	96.1	9.8	58.5	312.6
Sm	2.5	3.0	10.8	16.7	17.6	10.0	17.3	12.7	14.6	15.3	16.8	4.2	18.8	64.3
Eu	1.0	0.6	4.1	3.9	4.8	3.9	4.5	4.2	4.7	4.8	4.7	2.2	8.6	21.2
Gd	2.4	2.8	9.9	14.8	15.6	9.6	15.7	12.0	12.1	12.8	12.9	8.3	33.7	67.4
Tb	0.3	0.4	1.4	2.3	2.3	1.4	2.2	1.6	1.8	1.8	1.8	1.7	6.4	11.5
Dy	1.5	2.4	7.0	12.1	11.9	7.2	11.5	8.2	9.4	9.4	8.7	11.7	34.5	71.2
Ho	0.3	0.5	1.4	2.3	2.1	1.3	2.1	1.5	1.7	1.7	1.6	2.5	5.7	12.7
Er	0.8	1.7	3.8	6.3	5.5	3.7	5.8	4.0	4.6	4.7	4.1	7.4	13.6	32.6
Tm	0.1	0.3	0.5	0.8	0.7	0.5	0.8	0.5	0.6	0.7	0.6	0.9	1.6	4.0
Yb	0.6	1.8	2.9	5.2	4.0	2.9	4.2	3.0	4.1	4.6	3.8	4.7	8.5	21.7
Lu	0.1	0.3	0.4	0.8	0.5	0.4	0.7	0.4	0.6	0.7	0.6	0.6	1.1	2.6
La/Yb	25	10	32	16	13	18	13	39	19	17	33	3	12	22

Hf, Nb, Ta and Th may be low due to incomplete digestion