Ba-Mg isotopes in high Ba-Sr granites record melt-metasomatized mantle and crustal growth

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Table S1 Mg isotope compositions of Caledonian high Ba-Sr granitoids from Scotland

Sample	Location	Lithology	$\delta^{25}Mg$	2SD	$\delta^{26}Mg$	2SD	N
RA1	Rogart	Appinite	-0.172	0.057	-0.328	0.014	3
RA2		Appinite	-0.146	0.014	-0.276	0.012	3
LPR		Appinite	-0.123	0.014	-0.247	0.012	3
RT1		Granodiorite	-0.113	0.015	-0.233	0.015	3
RHG2		Granodiorite	-0.123	0.025	-0.238	0.009	3
RHG2R		Granodiorite	-0.135	0.028	-0.266	0.028	3
RHG1		Granodiorite	-0.193	0.044	-0.370	0.025	3
R2		Granite	-0.219	0.012	-0.418	0.013	3
SR2	Strontian	Appinite	-0.128	0.037	-0.249	0.024	3
SR1		Granodiorite	-0.160	0.007	-0.295	0.030	3
SR3		Granodiorite	-0.154	0.007	-0.295	0.022	3
SR4		Granodiorite	-0.144	0.035	-0.270	0.016	3
SR4R		Granodiorite	-0.139	0.051	-0.279	0.035	3
BHVO-2			-0.104	0.014	-0.201	0.009	3
BIR-1			-0.101	0.050	-0.199	0.004	3
G-2			-0.072	0.017	-0.134	0.036	3
RGM-1			-0.112	0.010	-0.217	0.017	3

Footnotes:

1.the long-term external precision of $\delta^{26}Mg$ is better than 0.05‰ (2SD); 2.USGS reference materials, i.e.,-0.201 ± 0.009‰ for BHVO-2,-0.199 ± 0.004‰ for BIR-1, -0.134 ± 0.036‰ for G-2, and -0.217 ± 0.017‰ for RGM-1 are in good agreement with previously published

values from An et al. (2014)；3.There are two repeated samples (lable R), their results are also in good agreement with repeated values.

Table S2 Ba isotope compositions of Caledonian high Ba-Sr granitoids from Scotland

Samples	Location	Lithology	Ba(ppm)	$\delta^{137/134}$Ba	2SD	$\delta^{138/134}$Ba	2SD
Samples							
RA1	Rogart	Appinite	2620	-0.07	0.04	-0.10	0.05
RA2		Appinite	1590	-0.14	0.01	-0.17	0.00
LPR		Appinite	1897	-0.11	0.04	-0.13	0.03
RT1		Granodiorite	1357	-0.02	0.03	0.00	0.01
RHG2		Granodiorite	1093	0.00	0.02	0.03	0.02
RHG2R		Granodiorite	1093	0.00	0.05	0.01	0.05
RHG1		Granodiorite	1396	-0.16	0.03	-0.19	0.01
R2		Granite	1195	-0.13	0.01	-0.16	0.00
SR2	Strontian	Appinite	809	0.02	0.02	0.03	0.02
SR2R		Appinite	809	0.03	0.02	0.02	0.02
SR1		Granodiorite	1112	-0.04	0.05	-0.05	0.02
SR3		Granodiorite	1022	0.06	0.06	0.07	0.05
SR4		Granodiorite	959	0.17	0.02	0.23	0.00
SR4R		Granodiorite	959	0.16	0.02	0.21	0.03
Blank reference samples							
BLANK-1					0.09 ng		
BLANK-2					0.14 ng		
BLANK-3					3.20 ng		
USGS reference materials							
GSP-2				0.03	0.01	0.01	0.00

AGV-2	0.00	0.01	0.01	0.01
GBW07111	-0.02	0.04	-0.02	0.05
G-2	0.03	0.01	0.03	0.02

Footnotes: The external precision of $\delta^{138/134}$Ba based on the long-term measurements of the two inhouse standards (USTC-Ba and ICPUS-Ba) is better than 0.04‰ (2SD). We estimate the long-term external precision of $\delta^{138/134}$Ba is better than 0.05‰ (2SD), which has been verified by Deng et al. (2021).
The $\delta^{138/134}$Ba values yielded in this study for the two USGS reference materials, i.e., 0.01 ± 0.00‰ for GSP-2, 0.01 ± 0.01‰ for AGV-2, -0.02 ± 0.05‰ for GBW07111, and 0.03 ± 0.02‰ for G-2 are in good agreement with previously published values (Nan et al., 2015; Gou et al., 2020; Deng et al., 2021, 2022; An et al., 2020).

Table S3 Major and trace element and Sr-Nd isotopic data of high Ba-Sr granites from Scotland

Location	**Rogart**							**Strontian**			
Lithology	**Appinite**			**Granodiorite**			**Granite**	**Appinite**	**Granodiorite**		
Samples	**RA1**	**RA2**	**LPR**	**RT1**	**RHG2**	**RHG1**	**R2**	**SR2**	**SR1**	**SR3**	**SR4**
SiO2	47.57	57.2	48.97	62.98	67.04	69.24	71.2	48.34	62.81	63.23	63.83
TiO2	1.13	0.5	1.39	0.77	0.63	0.43	0.61	1.5	0.73	0.78	0.76
Al2O3	12.41	10.03	9.77	15.4	15.3	14.7	14.2	12.89	15.46	15.9	15.94
Fe2O3T	7.98	5.34	6.45	4.71	3.87	2.63	2.78	8.92	3.98	4.24	3.99
MnO	0.12	0.1	0.12	0.07	0.05	0.04	0.04	0.13	0.06	0.07	0.06
MgO	9.42	11.7	9.06	3.14	2.39	1.3	1.9	9.97	2.61	2.65	2.4
CaO	9.22	9.7	10.12	4.09	3.06	1.96	2.56	8.17	3.65	3.77	4.2
Na2O	3.41	2.96	2.12	4.88	4.89	5	5.13	2.57	4.33	4.78	4.68
K2O	2.69	1.1	4.12	3.29	3.23	3.89	2.15	2.65	3.74	2.79	2.57
P2O5	1.42	0.36	1.06	0.36	0.27	0.15	0.16	0.68	0.23	0.26	0.23
Total	99.3	99.8	98.6	100	101	101	100	99.9	98.9	99.1	99.3
87Sr/86S(i)	0.70630	0.70616	0.70571	0.70633	0.70631	0.70636	0.70673	0.70577	0.70527	0.70559	0.70554
143Nd/144Nd(i)	0.51206	0.51204	0.51191	0.51190	0.51189	0.51189	0.51189	0.51239	0.51238	0.51237	0.51244
εNd(t)	-0.4	-1.1	-3.8	-4.9	-4.9	-4.9	-4.3	0.6	0.6	0.1	1.3
K2O+Na2O	6.1	4.06	6.24	8.17	8.12	8.89	7.28	5.22	8.07	7.57	7.25
A/CNK	0.49	0.42	0.37	0.81	0.89	0.92	0.92	0.59	0.87	0.90	0.88
Li	42.23	15.10	60.08	17.76	18.29	11.10	10.70	19.32	21.61	18.88	33.49
Be	2.65	1.92	2.85	2.37	2.57	2.23	2.35	1.87	2.58	2.13	3.22
Sc	29.00	23.93	17.78	9.09	7.48	4.38	2.26	28.51	7.61	8.14	7.93
V	192.74	83.35	123.96	73.45	58.57	39.34	21.97	197	69.28	72	67.72
Cr	71.33	975.17	437.94	115.72	79.62	53.68	22.92	493	66.22	57	63.53

Co	45.23	50.93	40.16	45.97	38.33	71.75	69.07	59	99.16	82	63.88
Ni	69.74	219.76	93.51	59.99	41.00	28.76	12.69	205	46.25	44	38.67
Cu	62.32	23.12	17.44	2.86	2.59	2.71	2.26	46	16.80	10	9.92
Zn	72.19	56.23	107.06	74.97	64.83	42.43	29.05	81	54.46	56	53.21
Ga	17.88	14.19	17.38	21.80	21.81	19.39	19.26	20	20.19	21	20.66
Ge	1.86	1.30	1.57	0.92	0.70	0.64	0.51	1	0.84	1	0.80
Rb	70.9	53.2	132.4	69.2	90.2	78.0	67.4	46.0	57.4	49.2	56.2
Sr	1799	998	1782	1299	1061	678	1096	1068	1088	1119	1032
Y	34.33	18.73	28.04	18.12	16.50	10.58	7.14	33	14.26	14	15.36
Zr	158.49	127.02	252.70	69.64	212.94	77.19	123.73	165	80.70	218	108.65
Nb	13.50	7.73	24.95	13.01	13.88	8.95	5.29	14	13.71	12	15.37
Cs	2.20	0.55	9.11	1.55	2.45	2.17	0.93	1	1.17	1	1.75
Ba	2620	1590	1897	1357	1251	1396	1195	750	1112	1020	959
La	157.7	123.3	91.1	66.9	49.99	50.8	29.6	64.0	53.7	45.3	46.4
Ce	328.6	245.9	227.0	131.0	99.75	91.9	55.9	140.8	108.1	84.4	88.9
Pr	40.21	26.40	27.21	14.10	11.63	9.66	5.97	17.77	11.47	9.19	9.34
Nd	169.89	102.45	108.33	54.43	44.34	34.42	21.65	75.59	42.63	34.18	34.20
Sm	30.47	16.73	17.84	8.45	7.12	5.18	3.39	13.75	6.60	5.42	5.53
Eu	6.75	3.55	4.11	2.07	1.88	1.39	0.98	3.36	1.81	1.70	1.51
Gd	20.22	11.35	12.64	6.36	5.69	3.84	2.50	11.03	4.97	4.61	4.41
Tb	2.08	1.09	1.39	0.74	0.65	0.43	0.29	1.32	0.58	0.53	0.56
Dy	8.13	4.38	6.22	3.68	3.10	2.06	1.39	6.51	2.91	2.63	2.94
Ho	1.21	0.67	1.00	0.64	0.55	0.35	0.24	1.16	0.50	0.47	0.54
Er	3.10	1.75	2.62	1.76	1.56	0.97	0.65	3.14	1.37	1.29	1.46
Tm	0.36	0.21	0.32	0.23	0.21	0.13	0.09	0.41	0.19	0.17	0.21
Yb	2.05	1.29	1.91	1.45	1.37	0.83	0.57	2.50	1.17	1.09	1.34

Lu	0.29	0.19	0.27	0.20	0.20	0.11	0.09	0.35	0.17	0.16	0.19
Hf	4.31	3.34	5.72	1.82	5.24	2.16	3.55	4.43	2.10	4.83	2.78
Ta	0.87	0.60	1.27	1.22	1.33	1.56	0.73	0.78	1.76	1.31	1.80
Pb	26.37	23.40	29.63	17.13	24.94	23.03	22.85	10.02	20.88	16.73	14.83
Th	25.06	19.10	12.01	9.25	10.43	10.26	8.99	9.76	11.50	7.69	7.22
U	5.97	3.04	3.68	1.51	2.47	1.83	2.89	2.07	2.90	2.11	2.48
Rb/Ba	0.03	0.03	0.07	0.05	0.07	0.06	0.06	0.05	0.05	0.05	0.06
Rb/Sr	0.04	0.05	0.07	0.05	0.09	0.12	0.06	0.05	0.05	0.04	0.05
NaO2/K2O	1.27	2.69	0.51	1.48	1.51	1.29	2.39	0.97	1.16	1.71	1.82
La/Yb	76.9	95.5	47.8	46.1	40.7	61.5	51.6	28.9	45.9	46.4	34.6
Th/Nd	0.15	0.19	0.11	0.17	0.24	0.30	0.42	0.07	0.27	0.22	0.21
Ba/Th	104.6	83.3	157.9	146.7	119.9	136.1	132.9	161.8	96.7	511.0	132.9
Ba/La	16.61	12.90	20.82	20.28	25.00	27.47	40.35	11.91	20.71	20.99	20.68
Th/La	0.16	0.15	0.13	0.14	0.21	0.20	0.30	0.07	0.21	0.04	0.16
Th/Yb	12.21	14.79	6.30	6.37	7.60	12.42	15.66	2.13	9.83	1.90	5.39
Lu/Hf	0.07	0.06	0.05	0.11	0.04	0.05	0.02	0.08	0.08	0.03	0.07
CaO/Al2O3	0.74	0.97	1.04	0.27	0.20	0.13	0.18	0.63	0.24	0.24	0.26
La/Sm	5.18	7.37	5.11	7.91	7.22	9.80	8.72	5.80	8.14	8.76	8.39
Sm/Nd	0.18	0.16	0.16	0.16	0.17	0.15	0.16	0.17	0.15	0.16	0.16
Ce/Pb	12.46	10.51	7.66	7.65	4.00	3.99	2.45	14.05	5.18	5.05	5.99

Major and trace element reference from Fowler et al. (2008) and Fowler and Rollinson (2012) and this study. Trace element analysed at Northwest University, China and method referenced from Qi et al. (2000).

Table S4 Trace element data of blank and International Geological Reference Materials

Sample Name.	Blank ppb	BHVO-2	BHVO-2	Ref. BHVO-2	AGV-2	AGV-2	Ref. AGV-2	BCR-2	Ref. BCR-2	GSP-2	Ref. GSP-2
Li	0.0026	4.64	4.59	4.80	11.0	11.0	11.0	9.43	9.00	34.5	36.0
Be	0.0000	1.12	1.05	1.00	2.22	2.26	2.30	2.35	1.60	1.49	1.50
Sc	0.0000	31.6	31.5	32.0	13.2	13.2	13.0	33.6	33.0	6.65	6.30
V	0.0633	317	315	317	119	119	120	421	416	51.2	52.0
Cr	0.4377	278	296	280	18.4	19.2	17.0	17.2	18.0	22.5	20.0
Co	0.0000	44.5	43.8	45.0	15.9	16.1	16.0	37.6	37.0	7.81	7.30
Ni	0.0454	118	151	119	24.7	20.5	20.0	13.1	13.0	17.3	17.0
Cu	0.0128	126	127	127	52.6	52.2	53.0	21.1	21.0	44.3	43.0
Zn	0.1070	99.3	99.0	103	90.4	90.1	86.0	127	127	106	120
Ga	0.0008	21.3	21.1	21.7	21.0	20.8	20.0	22.3	23.0	24.8	22.0
Ge	0.0017	1.60	1.68	1.60	1.06	1.04		1.77		1.69	2.15
Rb	0.0108	9.83	9.73	9.11	66.1	66.0	66.3	46.2	46.9	233	245
Sr	0.0076	394	391	396	665	665	661	340	340	238	240
Y	0.0000	26.6	26.3	26.0	20.3	20.7	20.0	36.4	37.0	26.1	28.0
Zr	0.0039	170	169	172	232	235	230	187	184	120	550
Nb	0.0002	18.4	18.4	18.1	14.3	14.4	14.5	12.7	12.6	26.5	27.0
Cs	0.0001	0.096	0.10	0.10	1.16	1.16	1.16	1.13	1.10	1.17	1.20
Ba	0.1347	132	132	131	1129	1122	1130	666	677	1350	1340
La	0.0068	15.1	15.1	15.2	37.6	37.7	37.9	24.7	24.9	181	180
Ce	0.0162	37.2	37.4	37.5	69.4	70.0	68.6	52.5	52.9	439	410
Pr	0.0011	5.26	5.23	5.35	8.17	8.15	7.84	6.74	6.70	50.6	51.0
Nd	0.0029	25.1	25.3	24.5	32.0	31.8	30.5	29.6	28.7	195	200
Sm	0.0006	6.09	6.15	6.07	5.62	5.63	5.49	6.65	6.58	26.6	27.0

Eu	0.0001	1.93	1.95	2.07	1.63	1.64	1.54	1.94	1.96	2.36	2.30
Gd	0.0001	5.88	5.94	6.24	4.80	4.81	4.52	6.54	6.75	15.7	12.0
Tb	0.0001	0.92	0.93	0.92	0.65	0.65	0.64	1.04	1.07	1.51	1.31
Dy	0.0000	5.29	5.34	5.31	3.59	3.57	3.47	6.45	6.41	5.86	6.10
Ho	0.0001	0.96	0.96	0.98	0.67	0.67	0.65	1.29	1.28	0.93	1.00
Er	0.0001	2.51	2.52	2.54	1.85	1.85	1.81	3.64	3.66	2.46	2.20
Tm	0.0000	0.33	0.34	0.33	0.26	0.26	0.26	0.53	0.54	0.27	0.29
Yb	0.0000	1.96	2.01	2.00	1.65	1.66	1.62	3.31	3.38	1.43	1.60
Lu	0.0001	0.27	0.27	0.27	0.25	0.25	0.25	0.49	0.50	0.18	0.23
Hf	0.0000	4.33	4.34	4.36	5.17	5.14	5.00	4.82	4.90	3.17	14.0
Ta	0.0000	1.15	1.15	1.14	0.83	0.84	0.87	0.77	0.78	0.87	0.85
Pb	0.2614	1.78	1.45	1.60	12.6	12.6	13.2	9.64	11.0	43.3	42.0
Th	0.0029	1.19	1.20	1.22	6.09	6.08	6.10	5.79	5.70	99.0	105
U	0.0001	0.41	0.42	0.40	1.89	1.87	1.86	1.66	1.69	2.30	2.47

All International Geologic reference data from Jochum et al. (2016).

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