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Uranium in Metamorphic Rocks

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Abstract. The distribution of U has been studied in two metamorphic rock-series with a gradient of regional metamorphism. One series ranges from the lowest greenschist to amphibolite facies and the other one shows increasing metamorphic grade from amphibolite to granulite facies. Several medium and high pressure granulitic inclusions from alkali basalts were also analyzed. The abundances of U in the rocks do not appear to be affected by metamorphism below the granulite facies grade. Granulites are depleted in U in comparison with equivalent rocks of amphibolite facies grade. There are also differences in their U distribution, as the bulk of U in amphibolite facies rocks is located along the fractures and cleavage planes of ferro-magnesian minerals and in U-rich accessories, while in granulites, most of the U resides in accessory minerals. It seems that the depletion of U in granulites is due to a loss of U which is not located in accessory minerals or in the crystal structure of rock-forming minerals and may also be related to a migration of hydrous fluids, perhaps during dehydration.

Introduction

The behaviour of uranium during metamorphic processes is still not well known. Several recent studies (Lambert and Heier, 1968; Heier, 1973) have shown that granulite facies rocks are depleted in U compared to rocks of lower metamorphic grades. There are also indications that some U might be already lost at a moderate grade of progressive metamorphism (Heier and Adams, 1965; Lambert and Heier, 1967). However, the actual mechanism or reactions leading to the release of U during metamorphism are not well established and it has even been suggested that the low abundance of U in granulites is a primary feature, unrelated to metamorphism (Holland and Lambert, 1973; Dupuy et al., 1977). Furthermore, only a few data have been reported on the location of U in metamorphic rocks and whether the bulk of U is present in major rock-forming minerals, U-rich accessories or is concentrated along the grain boundaries, fractures etc. is not exactly known.

The purpose of this study is to present data on the abundances of U in rocks of different metamorphic grades and their constituent minerals, to compare the concentration of U in some of these rocks with that of several other elements (K, Rb, Ba, Zr, and Ti) and to evaluate the behaviour of U during regional metamorphic processes.

Two rock-series with progressively increasing grade of metamorphism, one ranging from the lowest greenschist to amphibolite facies and the other from amphibolite to granulite facies, have been analyzed together with several granulitic inclusions from alkali basalts.

Geological Notes and Sample Descriptions

The first series, the Devonian Littleton formation of New Hampshire, comprises pelitic rocks with a grade of metamorphism varying from below the lowest greenschist facies (fossiliferous shales) to amphibolite facies. The petrography and geochemistry of this progressively metamorphosed formation were described by Shaw (1954, 1956). According to their grade of metamorphism, Shaw (1954) subdivided the rocks into three groups. The low-grade rocks (mainly lower greenschist facies) include shales and slates. The medium-grade rocks (predominantly lower amphibolite facies) are schists which do not contain sillimanite or are not adjacent to sillimanite-bearing rocks and are usually composed of quartz, muscovite, biotite, staurolite and garnet and sometimes feldspars in subordinate amounts. The high-grade rocks (middle amphibolite facies) either contain sillimanite or are adjacent to sillimanite-bearing rocks. They are made up of quartz, muscovite, biotite and sillimanite with subordinate garnet and feldspars. The detailed geochemical study of Shaw (1954, 1956) has shown that the formation represents an isochemically metamorphosed sequence as far as the major and several trace elements are concerned.

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The second set of samples was collected in the valley of the river Strona (Valle Strona) in the Western Italian Alps. The extensively studied metamorphic sequence of the Valle Strona (Bertolani, 1968; Mehnert, 1975; Rivalenti, 1966; Sighinolfi, 1969) is a part of the Ivrea Zone and consists of alternating bands of metasedimentary and mafic metaigneous rocks with a well defined metamorphic gradient ranging from amphibolite to granulite facies. The amphibolite facies section comprises graphite-bearing biotite gneisses and striped amphibolites. Biotite gneisses are made up of quartz, K-feldspar, plagioclase and subordinate garnet and sillimanite, while amphibolites consist predominantly of amphibole and plagioclase. The granulite facies sequence is also composed of mafic metaigneous and metasedimentary ("acid") rocks (Mehnert, 1975). The "acid" granulites include narrow banded types (stronalites) containing predominantly quartz, plagioclase, perthitic K-feldspar and garnet and rocks with migmatitic character. Mafic granulites are composed mainly of pyroxene and plagioclase, sometimes with garnet and/or amphibole. Mehnert (1975) and Sighinolfi and Gorgoni (1978) have argued that the amphibolite-granulite sequence of the Valle Strona was formed by a progressive regional metamorphism of the same formations with a relatively uniform pre-metamorphic composition. Thus "acid" and mafic granulites are probably the higher metamorphic grade equivalents of biotite gneisses and amphibolites, respectively.

Granulitic inclusions are from the Neogene basaltic pipe of Bournac (Velay), Massif Central, France (Leyreloup, 1973, 1974). The petrography and geochemistry of the inclusions were given by Leyreloup et al. (1977) who have shown that the rocks have compositions similar to tholeiitic basalts. Some inclusions represent medium pressure granulites while others contain high pressure granulitic assemblages corresponding to a depth of about 25 km (Leyreloup et al., 1977).

Analytical Methods

K, Rb, Ba, Ti, and Zr were determined by X-ray fluorescence while the analyses of U were done by a fission track technique using Lexan detectors and a neutron dose $\sim 10^{17}$ n/cm^2. Standard glasses of 0.33 ppm U were used for calibration. The uranium concentrations were determined from the measured fission track densities by comparing them in the samples and standards. U in the whole-rocks was determined in pellets of homogenized samples according to the method described by Fischer (1970). In each whole-rock pellet, about 5000 traces were counted. The distribution and concentration of U in the individual phases were studied on polished thin section by fission track mapping (Kleeman and Lovering, 1967). The precision and accuracy of the data can be judged from the replicate analyses of the standard rock BCR-1 (Table 1). The precision of the values for the mineral phases is lower (up to 40%). Each value for the major rock-forming minerals given in Table 2 is an average of the determinations of 10–20 grains. Due to the small size of the grains and the high concentrations, the precision of U data for accessory minerals is poor, with an error probably up to 80%.

Results

Whole Rocks

The average contents of U and other lithophile elements in 18 rocks from the Littleton formation, subdivided according to metamorphic grade into three groups (Shaw, 1954), are given in Table 1. The uranium concentration in the individual samples varies between 1.3 and 3.2 ppm (Appendix 1). The values are well within the range of the U abundances of North America gray and green shales (Adams and Weaver, 1958) and of recent pelagic sediments from the Pacific and Atlantic oceans (c.f. Rogers and Adams, 1974). A comparison of the three groups of rocks with different metamorphic grades (Table 1) shows that there is no significant change in the abundances of U and other analyzed elements in pelitic rocks with increase of the metamorphic grade up to the amphibolite facies.

Fourty-one amphibolite facies rocks and thirty-five granulites analyzed from the Valle Strona include both metaigneous (mafic) and metasedimentary ("acid") rocks. The average content of the analyzed elements in the rocks from the Valle Strona is given in Table 1. The average of U in the metasedimentary rocks of the amphibolite facies is very similar to that of the Littleton formation, although the individual samples have a greater range of U concentrations, varying from 0.92 to 3.4 ppm (Appendix 1).

Compared to amphibolite facies rocks, the "acid" granulites are depleted in U, Rb and K by factors of about 3, 2.6 and 1.5, respectively. The depletion of U, Rb and K in granulites is also accompanied by higher K/Rb and lower Rb/Ba and $U/K \times 10^4$ ratios. On the other hand, Zr appears to be slightly enriched in granulites compared to amphibolite facies rocks.

The U content of mafic rocks of amphibolite facies is within the range of the U abundances of amphibolites from the Canadian shield (Eade and Fahrig, 1973) and from Australia (Lambert and Heier, 1968). In comparison with amphibolites, mafic granulites are lower in U by a factor of about 2.4 and have a lower $U/K \times 10^4$ ratio. Granulites also have slightly lower contents of K and Rb although the difference is not statistically significant. Ti, Zr, and Ba do not seem to be affected by the increase of metamorphism. A depletion of K, Rb, and U in granulites similar to that from the Valle Strona has been reported from numerous high grade metamorphic areas (Lambert and Heier, 1968; Heier, 1973). The U content of high and medium pressure granulitic inclusions from Bournac varies in a narrow range from 0.07 to 0.12 ppm (Appendix 1).

The absence of a distinct correlation of U with K, Rb, Ba, Zr, and Ti and in fact also with all major elements (our unpublished data) in the analysed rock-series indicates that the bulk of U does not reside in the major rock-forming minerals.

Table 1. Average element concentrations and ratios in several suites of metamorphic rocks

		n	U	K	Rb	Ba	Zr	Ti	U/K × 10^4	K/Rb	Rb/Ba
Littleton	LG	7	2.15 (0.40)	3.42 (0.52)	162 (25)	550 (74)	220 (34)	0.56 (0.08)	0.63	211	0.29
	MG	5	2.13 (0.36)	3.59 (0.59)	163 (37)	555 (184)	222 (18)	0.57 (0.04)	0.59	220	0.29
	HG	6	2.24 (0.62)	3.84 (0.55)	170 (27)	637 (165)	187 (45)	0.67 (0.06)	0.58	226	0.27
Valle Strona mafic	AF	18	0.54 (0.20)	0.58 (0.24)	16 (11)	166 (109)	119 (46)	1.14 (0.39)	0.93	363	0.10
	GF	11	0.22 (0.16)	0.45 (0.25)	11 (9)	136 (91)	107 (49)	0.93 (0.39)	0.49	409	0.08
metasedimentary	AF	23	2.29 (0.68)	2.83 (0.89)	131 (48)	563 (227)	210 (80)	0.57 (0.18)	0.81	216	0.23
	GF	24	0.74 (0.36)	1.93 (1.01)	50 (35)	551 (340)	293 (166)	0.72 (0.43)	0.38	386	0.09
Bournac	MP	6	0.10 (0.02)								
	HP	2	0.11 (0.02)								
BCR-1			1.79 (0.20)	1.46 (0.05)	47 (2)	648 (15)	184 (7)	1.33 (0.02)			

K and Ti in wt%, U, Rb, Ba, and Zr in ppm; n = number of samples; values in brackets = standard deviation; grade of metamorphism: *LG* = low grade (mainly lower greenschist facies), *MG* = medium grade (lower amphibolite facies), *HG* = high grade (middle amphibolite facies), *AF* = amphibolite facies, *GF* = granulite facies, *MP* = medium pressure granulite facies, *HP* = high pressure granulite facies

Table 2. U concentrations (in ppb) of individual mineral phases of some metamorphic rocks

No.	Rock type		plg	kf	q	cpx	opx	am	bi	gr	s	ru	ap	sph	z	Σ
1	metasedimentary	AF	22	15	< 5				31	40					> 5000	1870
1	metasedimentary	AF	17						26	32			7300			1200
1	metasedimentary	AF	25	21	< 5				42				> 5000		6300	2470
1	metasedimentary	AF	21		< 5				44				> 5000			1700
1	metasedimentary	GF	27							42			> 5000	> 5000		650
1	metasedimentary	GF	5		< 5				22	7			> 4000	> 4000		552
1	metasedimentary	GF	24		3					65		> 5000			> 5000	132
1	metasedimentary	GF	22							52			> 5000		> 5000	825
1	mafic	AF	28					42						> 5000		360
1	mafic	AF	10					26						> 5000		352
1	mafic	AF	26			55		67	27			> 5000				213
1	mafic	AF	15					50						> 5000		659
1	mafic	GF	< 10				< 15						> 5000			20
1	mafic	GF	7			64	21						> 5000	> 5000		191
1	mafic	GF	12				30					7500	> 5000			400
1	mafic	GF	15				26							> 5000	> 5000	205
2	mafic	MP	10			33	14	15	30			9300	7600			80
2	mafic	MP	11			56	12	16				7200	8800			72
2	mafic	MP	8			22	11						6700			116
2	mafic	HP	11			16			23	46			4100			119
2	mafic	HP	10				11				19		5200			95

No. 1 = Valle Strona, No. 2 = Bournac pipe
AF = amphibolite facies, *GF* = granulite facies, *MP* = medium pressure granulite facies, *HP* = high pressure granulite facies
plg = plagioclase, *kf* = K-feldspar, *q* = quartz, *cpx* = clinopyroxene, *opx* = orthopyroxene, *am* = amphibole, *bi* = biotite, *gr* = garnet, *s* = spinel, *ru* = rutile, *ap* = apatite, *sph* = sphene, *z* = zircon, Σ = whole-rock (determination of the homogenized whole-rock pellets)

Minerals

In order to evaluate whether the distribution of U in the amphibolite facies rocks differs from that of granulites, several rocks of both facies were studied by fission track mapping. The abundances of U in the mineral phases of some representative metamorphic rocks are given in Table 2 and were determined in clear unfractured crystals. The homogeneous distribution of tracks and the uniformity of the U content of individual phases throughout a given sample suggest that these values represent U in solid-solution.

In amphibolite facies rocks, in general, the major rock-forming minerals have a very low concentration of U (Table 2) most of which is concentrated along the fractures, margins and cleavage planes of mainly ferromagnesian minerals (10–200 times higher in U than minerals) and in accessory minerals (apatite, zircon, sphene, rutile) with U contents in the order of several ppm (Table 2). In some rocks, opaque minerals (magnetite-ilmenite) also have a high content of U. Due to an inhomogneous and highly variable distribution of U along the fractures and cleavage planes, the mass balance calculations for U in these rocks can be at most only approximate. The rough estimates indicate that about 60–70% of U in metasedimentary rocks of amphibolite grade is located along the cleavage planes and fractures of biotite and to a lesser degree of garnet. The remaining U resides predominantly in accessory minerals, frequently enclosed in biotite. In amphibolites, the bulk of U is concentrated along the cleavage planes, fractures and margins mainly of amphibole (50–60%), and in accessory minerals.

The bulk of U in granulite facies rocks is present in accessory minerals (apatite, zircon, sphene, rutile). Compared to amphibolite facies rocks, the amount of U along the cleavage planes, fractures and grain boundaries is very small or frequently not even detectable. The U content of major rock-forming minerals is low (Table 2) and comparable to that of amphibolite facies rocks. Table 2 shows that the overall distribution of U in granulitic inclusions from the Bournac pipe is rather similar to that of granulites from the Valle Strona with the bulk of U residing in accessory minerals.

Discussion and Conclusions

The abundances of U in the rocks do not appear to be affected by metamorphism below the granulite facies grade. The granulites are, however, depleted in U in comparison with equivalent rocks of amphibolite facies grade. There is also a difference in the distribution of U in rocks of these two facies. In rocks of the amphibolite facies, the bulk of U is located along the fractures and cleavage planes of ferromagnesian minerals and in accessory minerals while in granulites, most of the U resides in accessories. Comparable rocks of both facies usually contain the same U-rich accessory minerals (zircon, apatite, sphene, rutile) and there are no indications that accessory minerals in granulites are more abundant or richer in U than those of amphibolite facies rocks. This suggests that the depletion of U in granulites is mainly due to an absence or a very low amount of U associated with the fractures and cleavage planes of minerals in the rocks of this facies.

The low content of U in granulites is frequently attributed to partial melting (e.g. Fyfe, 1973). Anatexis and removal of granitic melt can explain the simultaneous depletion of several lithophile elements such as K, Rb, Cs and U observed in many granulite facies regions. However, some mafic granulites of basaltic composition, which probably did not undergo partial melting and do not differ in their K and Rb abundances from equivalent amphibolites, are also depleted in U (c.f. Lambert and Heier, 1968). A possible explanation is that U which is not located in accessory minerals or in crystal structures of major rock-forming minerals is leached and removed by migrating hydrous fluids perhaps related to dehydration. Such a process may also explain the depletion of U in high grade "acid" or "intermediate" metamorphic rocks with "normal" K/Rb ratios reported from some areas (e.g. Kalsbeek, 1976). Furthermore, the loss of U in granulitic rocks by migrating fluid phases could be one of the reasons for the availability of U during the Precambrian when many major U deposits are known to have been formed.

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Appendix 1. Element concentrations in several suites of metamorphic rocks

	U	K	Rb	Ba	Zr	Ti
Littleton						
LG	2.21	4.03	190	484	187	0.47
	2.39	3.29	160	552	222	0.57
	2.49	3.23	147	547	241	0.64
	2.03	3.25	167	546	246	0.63
	1.58	4.23	196	683	185	0.57
	2.66	2.80	127	453	272	0.61
	1.70	3.10	146	583	190	0.42
MG	1.90	2.82	119	334	220	0.52
	2.51	3.55	155	496	203	0.60
	2.38	4.46	222	840	233	0.62
	2.22	3.70	166	525	208	0.58
	1.64	3.42	152	582	248	0.53
HG	1.26	3.27	148	632		
	2.34	3.83	163	699	148	0.62
	3.16	4.08	179	587	241	0.71
	2.38	3.66	170	412	133	0.59
	1.96	4.80	218	911	206	0.74
	2.36	3.41	145	580	205	0.69
Valle Strona						
Mafic rocks						
AF	0.651	0.40	11	128	97	0.78
	0.360	0.27	6	184	85	0.99
	0.352	0.52	7	61	107	1.07
	0.213	0.22	5	40	82	0.87
	0.831	0.73	16	281	149	0.85
	0.659	0.81	27	412	127	0.97
	0.616	0.46	11	134	67	0.61
	0.396	0.62	10	42	115	1.01
	0.720	0.38	9	205	99	1.16
	0.372	0.71	18	204	92	0.99
	0.404	0.87	34	306	78	0.93
	0.983	0.32	9	53	181	1.64
	0.612	0.20	5	59	167	1.35
	0.488	0.88	39	137	47	0.99
	0.397	0.85	27	174	125	1.15
	0.545	0.66	17	117	111	1.07
	0.741	0.61	15	112	190	2.16
	0.350	0.90	29	336	219	1.89
GF	0.205	0.62	23	96	112	0.61
	0.400	1.05	29	372	78	0.69
	0.191	0.56	10	88	112	0.93
	0.080	0.27	4	216	190	0.84
	0.086	0.25	4	169	106	1.00
	0.236	0.61	16	146	96	0.71
	0.472	0.42	9	93	182	1.67
	0.132	0.32	10	100	49	1.64
	0.020	0.19	3	73	61	0.54
	0.503	0.25	4	54	141	0.98
	0.136	0.38	6	90	47	0.59
Metasedimentary rocks						
AF	2.55	3.72	158	1085	180	0.67
	3.01	3.15	178	462	239	0.58
	1.80	3.87	179	511	86	0.14
	1.83	2.99	134	753	195	0.48

Appendix 1 (continued)

	U	K	Rb	Ba	Zr	Ti
	0.922	3.10	110	856	275	0.32
	1.81	2.96	111	286	266	0.50
	2.47	2.57	116	734	238	0.60
	1.77	4.13	204	605	141	0.80
	1.70	3.41	192	529	236	0.79
	1.45	2.97	148	720	145	0.62
	1.87	4.76	219	854	209	0.77
	1.20	1.65	82	476	187	0.70
	1.86	1.87	102	318	234	0.50
	2.89	2.94	137	382	206	0.67
	2.21	2.65	98	492	178	0.45
	2.90	2.72	144	529	226	0.73
	3.05	2.94	122	448	237	0.68
	3.43	2.16	109	387	169	0.61
	3.07	1.24	29	700	31	0.15
	2.48	1.64	66	262	373	0.52
	2.76	1.39	52	244	409	0.50
	2.51	3.64	167	398	176	0.73
	3.08	2.69	157	915	190	0.59
GF	1.24	1.34	44	388	269	0.45
	0.555	3.25	84	909	189	0.28
	0.512	1.30	32	437	157	0.38
	1.40	2.98	82	749	156	0.66
	0.552	2.30	87	505	132	1.04
	0.825	3.58	88	883	93	0.40
	0.132	0.95	18	244	84	0.05
	0.627	4.26	122	528	110	0.14
	0.847	1.74	25	634	366	0.52
	1.32	3.00	86	784	259	0.91
	0.298	2.57	86	1219	238	0.61
	0.895	1.91	61	517	318	0.77
	1.04	0.97	14	136	587	1.03
	0.756	1.18	25	574	298	0.57
	0.519	0.88	12	165	449	0.76
	0.540	2.63	42	1449	278	0.79
	0.734	0.61	< 1	54	386	0.74
	0.827	1.10	14	793	552	1.30
	0.088	2.75	105	331	39	0.11
	0.833	2.29	56	592	238	0.61
	1.26	1.75	60	467	609	1.91
	0.650	1.01	10	175	333	1.38
	0.328	0.91	12	245	562	0.86
	1.07	1.16	37	452	328	0.97
Bournac						
MP	0.072					
	0.124					
	0.116					
	0.122					
	0.080					
	0.075					
HP	0.095					
	0.117					

K and Ti in wt %; U, Rb, Ba, and Zr in ppm. Symbols are same as in Table 1

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